Use of Greenlandic resources for the production of bricks

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Preface

This PhD thesis contains the results of my research, which was conducted at the Arctic Technology Centre, Department of Civil engineering at the Technical University of Denmark (DTU). The PhD study was co-funded by the Government of Greenland, The Danish Agency for Science, Technology and Innovation and DTU Civil Engineering. The main supervisor was Lisbeth M. Ottosen (Associated Professor, DTU Civil Engineering) and co-supervisors were Gunvor M. Kirkelund (Researcher, DTU Civil Engineering), Pernille E. Jensen (Associate Professor, DTU Civil Engineering) and Arne Villumsen (former head of the Arctic Technology Centre, DTU Civil Engineering).

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In August 2011 I had the pleasure of visiting the brickworks Petersen Tegl A/S, Denmark. I am very grateful to Erich Mick, Kim Reinecker and Christian Petersen for their helpfulness and support throughout my stay. Furthermore, Erich Mick is thanked for helping with transport logistics and for prompt reply to questions during the continuation of my studies.

Tanbreez Mining Greenland A/S is thanked for providing tailings samples from the Tanbreez project in South Greenland.

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At last but by no means least, a very grateful thank you to my wonderful and understanding husband, children and family – without your love and support none of this would have been possible!

Abstract

The housing situation in Greenland is critical. In 2012, approximately 20 % of the housing was dilapidated, and due to an increasing population in the larger towns, there is currently an urgent need for the construction of new homes. Today, materials used in the Greenlandic construction industry are mostly imported, and due to high import costs, it is therefore important to investigate the potential for local construction material production. The aim of this PhD-study was to identify and test suitable raw materials for such local production. Clay-based bricks were chosen due to several reasons:

- Bricks, in general, are durable and fire-resistant materials, which possess good thermal and acoustic properties. In the harsh Arctic to sub-arctic climate of Greenland, which places high demands on construction materials, especially the thermal properties and durability of bricks are desirable characteristics.
- Bricks are heterogeneous materials, which can accommodate materials of wide ranging compositions without losing their technical properties. For this reason, the ceramic industry has been identified as one of the major recipients for waste in the future. Wastes of different types are becoming an increasing environmental and logistical problem worldwide, and their recycling has therefore received much attention. The protection of the vulnerable Arctic nature is a great concern and waste management by ceramic incorporation should therefore be investigated.
- Historically, masonry constructions (including bricks) were dismissed in Greenland in the early 1950's, due to low frost resistance of the mortar. Although this dismissal might have been technically warranted at the time, the ceramic industry has since experienced extensive development, and today clay-based brick constructions can therefore be designed to withstand the Arctic climate conditions.
- In 2006, the Arctic Technology Centre, DTU Civil Engineering in collaboration with the Danish brickworks Petersen Tegl conducted a large scale brick pilot-production from Greenlandic glaciogene marine clay from an occurrence near the town of Kangerlussuaq. The bricks were used for the construction of a house in Sisimiut, Greenland and it was established that the Kangerlussuaq occurrence was suitable as raw material for bricks.

Large occurrences of fine-grained glaciogene marine sediments are, however, found throughout Greenland and the knowledge gaps identified and covered by this study are therefore:

- to establish the characteristics, similarities and variations of Greenlandic marine sediment occurrences in general, in respect to potential use as brick clays.
- to describe how the variations in the sediments influence the properties of produced bricks.
- to investigate the link between raw material characteristics, processing details and resulting technical characteristics of bricks produced from a representative sediment occurrence.

Furthermore, in relation to the potential for incorporating waste in the produced bricks, the following knowledge gap was identified and covered:

- to identify and conduct initial testing on waste types, which could be of interest in Greenlandic brick production. The waste types investigated in this study were municipal solid waste incineration (MSWI) ashes and tailings from the mining industry.

The major conclusions obtained from this study were:

- The Greenlandic marine sediments have very similar grain size distributions, mineralogy and major element chemistry. Furthermore, these properties are comparable to those of North American and North Scandinavian marine clays.
- Bricks with acceptable properties of e.g. compression strength, open porosity and water absorption can be produced from the sediments. However, the properties largely depend on the processing of the sediment, e.g. firing procedure and initial treatments.
- Addition of waste materials, such as the investigated bottom ash and mine tailings, generally improve the technical properties of the bricks, e.g. lowers the open porosity and increases the density. However, increased leaching of heavy metals was observed after firing, which could pose an environmental concern and requires further investigation.

Sammenfatning

Boligsituationen i Grønland er kritisk. I 2012 blev cirka 20% af de grønlandske lejligheder og huse erklæret saneringsmodne, og da befolkningen samtidig er stigende i de større byer, er der i øjeblikket et presserende behov for opførelse af nye boliger. I dag bliver materialer, der anvendes i den grønlandske byggebranche, hovedsageligt importeret, og på grund af høje import omkostninger, er det derfor vigtigt at undersøge potentialet for lokal byggematerialeproduktion. Formålet med dette PhD-studie var at identificere og teste egnede råmaterialer til en sådan lokal produktion. Mursten blev valgt som byggemateriale af flere årsager:

- Mursten er generelt holdbare og brandhæmmende materialer, som besidder gode termiske og akustiske egenskaber. Det arktiske til subarktiske klima i Grønland, stiller høje krav til byggematerialer, og her er især de termiske egenskaber og holdbarheden vigtige.
- Mursten er heterogene materialer, som er i stand til at inkorporere råmaterialer af meget forskellig art, uden at miste deres tekniske egenskaber. Af samme grund er den keramiske industri blevet identificeret som en af de største aftagere af affald i fremtiden. Affald er generelt et stigende miljømæssigt og logistisk problem i hele verden, og genanvendelse har derfor fået megen opmærksomhed. Beskyttelsen af den sårbare arktiske natur er af stor bekymring, og affaldshåndtering i form af keramisk inkorporering bør derfor undersøges.
- Historisk set blev murværk (inkl. mursten) erklæret uegnet til konstruktioner i Grønland i begyndelsen af 1950'erne, på grund af lav frostbestandighed af mørtel. Selv om der måske har været en teknisk berettigelse for dette på daværende tidspunkt, har den keramiske industri siden oplevet stor udvikling og murværk bør derfor i dag være særdeles egnet selv under arktiske klimaforhold.
- I 2006 gennemførte Center for Arktisk Teknologi, DTU Byg i samarbejde med det danske teglværk Petersen Tegl en større pilot-produktion af mursten lavet af grønlandsk glaciogent marint ler fra en forekomst nær Kangerlussuaq. Murstenene blev efterfølgende brugt til opførelse af et hus i Sisimiut og det blev fastslået, at Kangerlussuaq forekomsten var egnet som råmateriale til mursten.

Store forekomster af finkornede glaciogene marine sedimenter findes dog i hele Grønland og formålet med dette PhD-studie har derfor været:

- at fastlægge de generelle karakteristika, ligheder og variationer af grønlandske marine sediment forekomster med hensyn til potentiel anvendelse som teglværksler.
- at beskrive, hvordan variationer i sedimenterne påvirker egenskaberne af mursten.
- at undersøge sammenhængen mellem råmaterialeegenskaber, forarbejdning og tekniske egenskaber af mursten fremstillet fra en repræsentativ sediment forekomst.

Derudover er undersøgelser omkring inkorporering af affald i de producerede mursten også blevet udført med følgende formål:

- at identificere og gennemføre indledende test på affaldstyper, som kunne være af interesse for grønlandsk murstensproduktion. De undersøgte affaldstyper i dette studie var forbrændingsasker og affald (tailings) fra mineindustrien.

Hovedkonklusionerne fra PhD-studiet er:

- De grønlandske marine sedimenter har meget ens kornstørrelsesfordelinger, mineralogi og hovedelement kemi. Desuden er disse karakteristika sammenlignelige med tilsvarende for nordamerikanske og skandinaviske marine lerer.
- Mursten med gode egenskaber, såsom høj trykstyrke, lav åben porøsitet og lav vandoptagelse kan produceres fra de marine sedimenter. Imidlertid afhænger egenskaberne i høj grad af processeringen af sedimentet, f.eks brændingsproceduren og forarbejdningen.
- Tilsætning af affaldsmaterialer, såsom de undersøgte slagger og mineaffald, kan generelt forbedre de tekniske egenskaber af murstenene og f.eks. reducere den åbne porøsitet og øge densiteten. Der blev imidlertid observeret øget udvaskning af tungmetaller efter brændingen, hvilket kan have miljømæssige konsekvenser og derfor bør området undersøges yderligere.

Eqikkaaneq

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Contents

Introduction

Greenland

Greenland is the world's largest non-continental island covering an area of 2,166,086 km^2 whereof approximately 20 % is ice-free (Figure 1). Greenland is part of the Kingdom of Denmark and was granted home-rule in 1979 followed by Self-government in 2009. The population consists of 56,282 inhabitants (Jan., 2014) of which 67 % live in the 7 largest towns (Nuuk, Sisimiut, Ilulissat, Qaqortoq, Aasiaat, Maniitsoq and Tasiilaq) and the rest are distributed between smaller towns and settlements (Statistics Greenland, 2014 a).

Figure 1. Greenland (divided according to the four municipalities and the national park). Statistics Greenland (2014 a).

The towns and settlements are predominantly distributed along the West coast and the only means of transportation between them are by sea or by air. Transportation by sea is monopolised

by the government-owned company Royal Arctic Line A/S, which has liner shipping to Denmark and Iceland, as the only destinations outside Greenland. Today, Greenland is to a large extent reliant on import of e.g. food supplies and goods for construction. Most of these goods are imported as sea cargo from Denmark and Europe (Statistics Greenland, 2014 b).

Historical background on housing

Prior to the 1950's, homes in Greenland had mainly been self-build houses which generally did not meet the Danish mid-twentieth century requirements for health and safety. In 1950 the Greenlandic commission under the Danish government presented a report on the general situation in Greenland and recommendations for improvements. The commission concluded that Greenlandic homes were generally too small, cold and badly insulated and recommended that new pre-designed homes, constructed from imported pre-fabricated materials, should begin to replace the old ones (Grønlandskommissionens betænkning, 1950). As a result of the commission's report, the Danish government introduced new regulations in Greenland, where all homes had to be constructed by skilled labourers. Furthermore, a small range of pre-designed house-types, were favourably subsidised by the government, which meant that these buildings and their choice of materials became dominant in Greenland. From 1956 until the early 1970's approximately 10 types of pre-designed houses were subsidised (Madsen, 2000).

Historical background on bricks

In the commission's report a single paragraph discussed the building materials, which were to be used for the construction of the new homes. The report stated: *'During the discussions, the commission was informed that brick houses were not suitable for the Greenlandic climate, because the mortar is not able to withstand the large temperature variations, mainly due to the effects of frost'* (Grønlandskommissionens betænkning, 1950). Although wood had several disadvantages (such as low fire-resistance and high maintenance requirements) it was preferred because of its durability during transport, good insulation properties and because maintenance could be carried out by unskilled labourers. Furthermore, concrete was deemed suitable although it was expensive.

Although, the commission's recommendations of excluding bricks as a major building material might have been warranted at the time, extensive research has since led to improved production methods, standardisations and codes of practice for masonry constructions (Davison, 1975;

Grimm, 1985; Hendry and Khalaf, 2001; Melander and Ghosh, 1996). Brick houses are today considered suitable even in extreme climates (Brick Industry Association, 2006 a). Furthermore, the steady low temperatures, which prevail during the winters in many parts of Greenland, are less likely to destroy the bricks and mortar; On the contrary, exposure to humid climates and frequent freeze-thaw cycles are much more demanding and make porous materials, such as bricks and mortar, more susceptible to damage (Taylor, 1991).

Bricks have several other advantages that make them an interesting building material in arctic climate regions, such as Greenland. They are versatile materials, which, depending on the raw materials and production methods, can develop specific properties. Highly sintered bricks are often very durable, due to their low water absorption, low porosity and high strength. These types of bricks are suited as facing bricks, i.e. bricks subjected to outdoor climate. Bricks with high porosity have excellent thermal insulation properties (Dondi, 2004) which make them suitable for indoor use. In general, bricks require almost no maintenance in comparison to e.g. wood, have good acoustic properties and are fire-resistant (Hornbostel, 1991). In spite of this, wood and concrete are still favoured building materials in Greenland today (Bjarløv and Vladykova, 2011; Garcia; 2012; Madsen, 2000) and only a very limited number of bricks have been used for the construction of houses and chimneys.

Future challenges

Today, the population is increasing in the Greenlandic towns and the present housing situation is critical, due to long waiting lists and poor conditions of the current buildings (Garcia, 2012; Naalakkersuisut, 2012). In 2012 the government of Greenland reported that about 70 % of the state-owned flats and terraced houses, which constitutes approximately 20 % of the housing in Greenland, were dilapidated (Naalakkersuisut, 2012; Statistics Greenland, 2010). In order to accommodate an increasing population and satisfy the demands for housing, it is therefore vital for the government of Greenland to construct new homes for the future. However, as most of the construction materials used in the country today, are imported, it is of great importance to investigate the potential for local production.

The raw materials for local production of construction materials could be either natural or secondary, i.e. waste materials. In respect to waste materials, especially the managing of

hazardous waste is a challenge in the small Greenlandic society. Today hazardous waste from households and commercial industries is collected and exported to Europe (Eisted and Christensen, 2011). On an industrial basis, the amount of hazardous waste is likely to increase if large scale mining operations commence. Greenland has recently experienced increased interest from mineral exploration companies and although a recent report (the Committee for Greenlandic Mineral Resources to the Benefit of Society, 2014) questioned the extent of future mining operations in Greenland, it is very likely that large-scale operations will begin within 5-10 years. Most of these operations will produce potentially hazardous bi-products (tailings) and at the same time demand construction materials for new housing and infrastructure etc.

Previous investigations and research aims

In relation to the above described issues, the overall research aim of this PhD project was therefore to investigate the technical potential for local brick production in Greenland. The idea of producing bricks from local Greenlandic resources is, however, not new. Prior to this PhD project preliminary investigations had been conducted on fine-grained marine glaciogene sediment occurrences in West Greenland. As these studies were all reported in Danish, a brief summary of the investigations are given here.

In 1986 the company A/S Carl Nielsen, on behalf of the municipality of Qasigiannguit (Qasigiannguit kommunea), conducted a short survey of a large occurrence south of Qasigiannguit, West Greenland (Kalvig , 1990). Fired specimens were prepared in order to study shrinkage, colour and sintering. Although properties such as strength, density and water absorption of the fired specimens were not tested, the study concluded that there was a good potential for brick and tile production. Efflorescence was observed on the specimens after firing.

In 2002 the company NunaMinerals A/S, on behalf of the municipality of Nuuk (Nuup kommunea), conducted a survey of a large occurrence near Nuuk (NunaMinerals A/S, 2002). Based on initial sediment characteristics, it was concluded that bricks could likely be produced from this sediment. Fired specimens were, however, not prepared (or at least not reported) in order to confirm this. It was estimated that the investigated occurrence contained > 2 million m^3 of sediment, which would correspond to approximately 1 billion bricks. For comparison, it was estimated that the annual production of a Danish brickworks was between 20-60 million bricks.

In 2001 the Arctic Technology Centre, DTU Civil engineering, initiated the investigations on use of an occurrence near the town of Kangerlussuaq for the production of construction materials, such

as bricks, expanded clay aggregates and as cement-replacing filler. Several student projects were conducted in order to establish the material properties, extent of the clay resource and its potential usage in the respective construction materials (summarised in Appendix 4). The preliminary investigations showed a promising potential as brick clay, although the salt content of the clay was very high. In 2006 a larger scale pilot production of bricks was conducted in collaboration with the Danish brickworks Petersen Tegl A/S. The processing details of the brick production were, however, not recorded. The produced bricks were used in the construction of a community centre in the Greenlandic town of Sisimiut in 2007. The building is still in use and in good condition (Villumsen, 2013).

As the preliminary investigations on the marine glaciogene sediment occurrences had demonstrated promising potential, it was decided to continue investigations on these materials in the present study. In relation to the previous investigations and pilot productions, the following three knowledge gaps of major importance were identified:

- How similar are the marine sediment occurrences in Greenland? Although marine glaciogene sediment occurrences are distributed throughout Greenland, their general characteristics, similarities and variations in respect to potential use as brick clays are not well known.
- How do the variations in the sediments influence the properties of produced bricks? As a continuation of the above described knowledge gap, it is important to conduct a screening where the raw material and some resulting properties after firing can be assessed.

Addressing the first two knowledge gaps are essential in order to assess whether or not marine glaciogene sediments in general are suitable as raw materials for brick production.

- What are the technical characteristics of bricks produced from a representative occurrence? This step should include the production of test-bricks, in order to study the link between the raw material characteristics, processing details and resulting technical characteristics of the produced bricks. This link has not been addressed in previous studies, but is important in order to evaluate the suitability of the raw material and to assess the technical potential of the produced bricks.

Besides the marine sediment occurrences, local waste materials were chosen as secondary raw materials for incorporation in the bricks. Clay-based ceramics, such as bricks, are heterogeneous materials, which can incorporate raw materials of wide ranging compositions, without impairing

their technical properties (Dondi et al., 1997a,b; Segadães et al., 2005; Torres et al., 2009 b). Due to this ability, bricks have become a popular material in waste management research worldwide and several studies have demonstrated that clay-based bricks and tiles can successfully accommodate waste types, such as incineration ashes, mine tailings and dredged harbour sediments. Table 1 lists a selection of some of the more recent studies with respect to the variations in tested waste types and their reported benefits.

Waste type	Reference	Summary and conclusions	Role of waste	Benefits/disadvantages of use
Mine tailings and quarry residues				
• Gold mill tailings	Roy et al. (2007)	5-75% of gold mill tailings were mixed with two types of clayey soils and fired at temperatures of 750-950 °C. Bricks containing between 45-75% mill tailings fulfilled criteria for compressive strength, shrinkage and water absorption.	Inert material	Benefits: • Lowered costs compared to traditional bricks • Solving tailings disposal problems
• Granite wastes	Menezes et al. (2005)	20-60 % of granite sawing wastes were mixed with a red brick clay and fired at temperatures in the range of 800-1000°C. Bricks incorporating 20-50 % of granite sawing waste met the requirements for water absorption and modulus of rupture.	Clay replacement	Benefits: • Reduced shrinkage Disadvantages: • higher firing temperature to obtain sintering
• Hematite tailings	Chen et al. (2011)	77-100 % of hematite tailings were mixed with 0-15 % brick clay and 0-8 % fly ash and fired at 1000°C. The optimum composition contained 84 % hematite tailings and conformed to Chinese standards.	Clay replacement	Benefits: • Natural resource conservation • Solving tailings disposal problems
Other industrial residues				
• River silt from silica sand washing	Torres et al. (2009 b)	5-10% of river silt (RS) were mixed with brick clays and fired at 950-1100°C. Incorporation of up to 5 % of RS did not change the final properties.	Clay replacement (of low plasticity clay)	Benefits: • Avoiding negative environmental impact • Lowered production cost of ceramics

Incineration ashes				
• Municipal solid waste incineration (MSWI) bottom ash	Kasuriya et al. (2008)	20-60 % of MSWI bottom ash were mixed with a brick clay mixture and fired at temperatures in the range of 1000-1125°C. Addition of 40-60% bottom ash improved the flexural bending strength and lowered water absorption.	Clay replacement	Benefits: • Improved sintering • Solving disposal problems
• Municipal solid waste incineration (MSWI) fly ash	Zhang et al. (2011)	15-30 % of MSWI fly ash were mixed with 50-65 % brick clay and 5-15 % feldspar and fired at temperatures of 900-1050°C (orthogonal test). Addition of 20 % MSWI fly ash and firing at 950-1000°C was optimal for brick production.	Clay replacement or inert material	Benefits: • Reduced heavy metal leaching after firing
Other				
• Cigarette butts	Kadir and Mohajerani (2008)	2.5-10 % of cigarette butts were mixed with brick clay and fired at 1050°C. Addition of 2.5-10% of cigarette butts could be used for production light-weight bricks depending on requirements.	Additive	Benefits: • Avoiding negative environmental impact • An estimated reduction of 60% in firing energy when 5 % cigarette butts were added Disadvantages: • Reduced strength and density and increased porosity and water absorption with increasing amounts of cigarette butts
• Dredged harbour sediment	Mezencevova et al. (2012)	50-100% dredged materials were mixed with a brick clay and fired at temperatures of 900-1000°C. A composition of 50% dredged material and 50% brick clay fired at 1000° proved to be favourable, due to good compressive strength and resistance to freeze-thaw cycles.	Clay replacement	Benefits: • Solving disposal problems • Natural resource conservation

Table 1. A selection of studies on waste utilisation in clay-based ceramics.

The benefits of waste addition, which are most often mentioned in the literature, are:

1) conservation of natural resources, e.g. by replacing natural clay with waste.

2) improved technical properties of the final product, e.g. improved mechanical strength.

3) lowering of energy consumption in production, i.e. by lowering the firing temperature.

4) cost reductions on the final product, e.g. due to lower costs on raw materials.

5) solving disposal problems and protecting the environment.

In Greenland, only a small number of studies have investigated the potential for using Greenlandic waste, such as municipal solid waste incineration (MSWI) bottom ash and contaminated harbour sediments for road construction (Appendix 7; Kirkelund et al.,2012) or fly ash for replacing cement (Juel Olsen and Westmose Hansen, 2013). As bricks are neither used to a great extent nor produced locally, the incorporation of Greenlandic wastes into bricks has not previously been studied. The important knowledge gaps identified in this connection were:

- What waste types could be of interest in Greenlandic bricks? How would they influence the properties of produced bricks? This step should include a screening on selected local waste materials for incorporation in bricks as partly substituting for local clay.

Summary of knowledge gaps and objectives and structure of the thesis

The identified knowledge gaps and objectives are listed in Table 2. Each knowledge gap has been assigned a letter (A-D) and studies related to the individual knowledge gaps are listed according to appendix number.

	Knowledge gap	Objective	Appendix
A	How similar are the marine sediment occurrences in Greenland?	To make a general characterisation of marine sediment occurrences in West Greenland.	Appendix 1, Appendix 2
B	How do the variations in the sediments influence the properties of produced bricks?	To conduct a screening of selected occurrences, in order to determine properties, which can be compared to properties of brick clays from other parts of the world.	Appendix 2
C	What are the technical characteristics of bricks produced from a representative occurrence?	To produce test-bricks from a suitable occurrence in order to determine relevant characteristics such as e.g. compression strength and water absorption.	Appendix 3, Appendix 4
D	What waste types could be of interest in Greenlandic bricks? How would they influence the properties of produced bricks?	To conduct a screening of local waste materials for incorporation in bricks.	Appendix 5, Appendix 6, Appendix 7

Table 2. Knowledge gaps and objectives of this PhD-study.

Research methodology

The research methodology is here described generally. Please refer to the relevant appendices for specific details on the individual studies. The overall methodology applied in this study is sketched in Figure 2. Marine sediments and the resulting brick properties were studied in order to obtain the best choice of available raw material. Once this was decided, different waste types were added to the raw material in order to study the effect of these on brick properties and to test whether the waste addition could optimise or benefit the produced bricks.

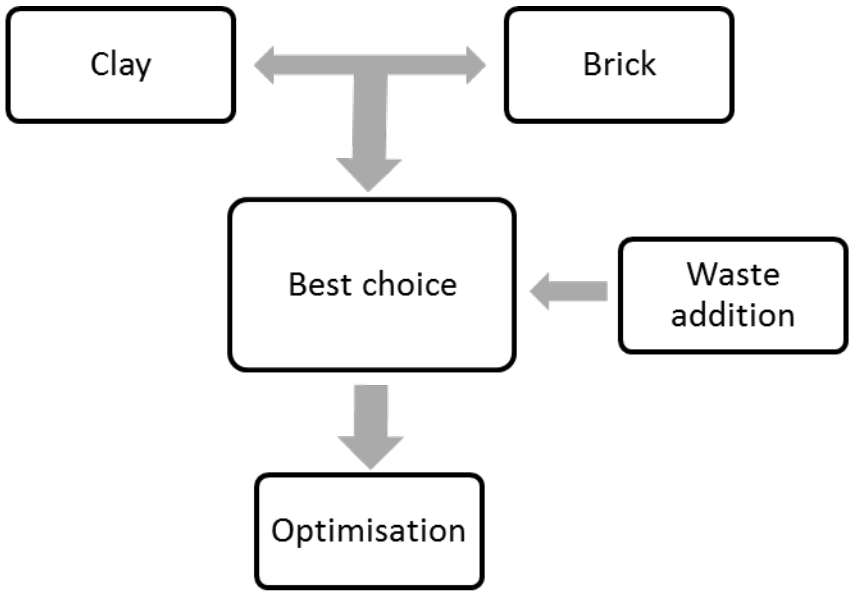

Figure 2. Sketch of the overall research methodology

Experimental procedure

The ceramic processing applied in this study is sketched in Figure 3. In the following, the different steps and their characteristic analyses and methods will be described briefly.

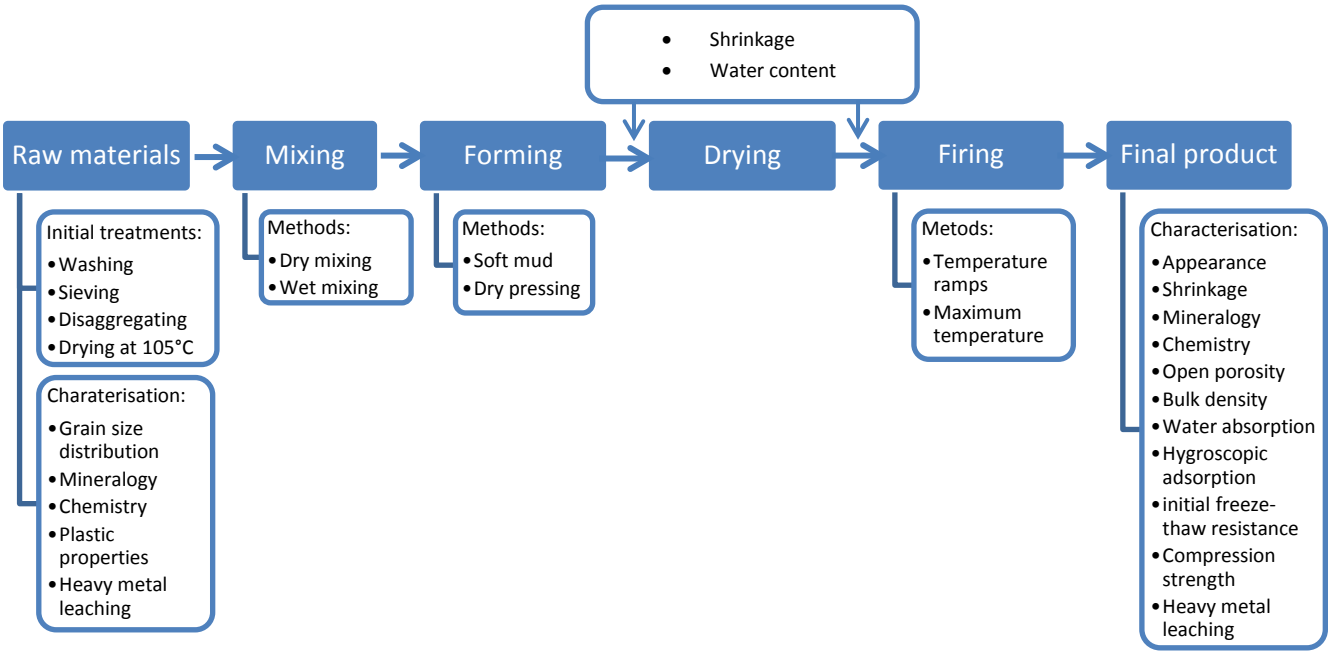

Figure 3. Flow chart of the ceramic processing (modified from Reed, 1995), characteristic analyses and methods applied during this study.

Raw materials

Initial characterisation is an important tool for gaining insights on the materials of interest and for identifying critical issues which should be addressed. As seen from Figure 3. five categories of characteristic analyses were employed during this study. The grain size distributions of the investigated materials are important as they determine the possible packing arrangement of the grains and ultimately properties such as strength, porosity and water absorption (Holmboe, 2001). When bricks are fired, the sintering occurs at the contacts between grains and in order to obtain a high strength it is therefore important to have as many contacting grains as possible. This is obtained by an even grain size distribution consisting of both smaller and larger grains (Holmboe, 2001). Two analytical methods for grain size analyses were employed: 1) sieving combined with the hydrometer method and 2) laser diffraction. Laser diffraction (dry-method) was chosen in the studies where waste materials were included (Appendix 3 and 5). This method was chosen, due to a high content of salt in the fly ashes, which would have dissolved during the use of the hydrometer analyses. For consistency, all the investigated waste materials were therefore analysed by laser diffraction.

The major element chemistry combined with mineralogy is a powerful tool for investigating and predicting the behaviour of the raw materials during firing. The ratio of $CaCO_3$ to Fe_2O_3 is determinative for the colouration of the fired product. Raw materials containing $CaCO_3$ and Fe_2O_3

in a ratio of 3:1 or higher obtain a yellow colour upon firing, whereas lower ratios give a red colour (Andersen et al., 1989). Furthermore, minerals such as quartz are important when deciding on the ramp rates of the firing programme. For quartz, the phase change from α to β structure at 573°C involves a volume change, which can develop internal stresses and lead to cracking if the temperature increase (or decrease) around this transition occurs to rapidly (García-Ten, 2010; Holmboe, 2001). The alkali metals and alkaline earth metals (Na, K, Mg, Ca) are known as fluxing materials and contribute to fusing of the particles (Bloodworth et al., 2007; Torres et al., 2009 a). In order to fully understand complex systems, it is often useful to employ an empirical approach and at the same time gather as much information about the system through characterisation. This type of classical approach has been applied in this study, due to the complexity of the clay and the other raw materials investigated here.

In some of the studies (Appendix 5, Appendix 6) initial treatments, such as sieving and disaggregation, of the raw materials were applied. Furthermore, heavy metal leaching tests were conducted on potentially hazardous waste materials.

Forming

In this study, two types of forming methods were employed: the soft mud method and the dry-pressing method (Brick Industry Association, 2006 b). The soft mud method is the traditional moulding-by-hand method and although it is seldom used in modern industrialised brickworks, it is very practical for small scale experiments such as the one applied in Appendix 3. In this study a water lubricated wooden mould was used for forming (dimensions: length: 18.6 cm, width: 12 cm, height: 6 cm) and a wooden piston was used to aid the removal of the mould from the formed samples (green bodies). The dry-pressing method is suitable for clays of low plasticity and involves the compaction of a granular powder into a mould by applying pressure (in this study the applied pressure was approximately 47 MPa). The water content of the powder is kept below its plastic limit. In this study, a mould was purpose-built for the production of brick-pellets, which were approximately 2 cm in diameter and 3-4 mm in height. The two methods are shown in Figure 4.

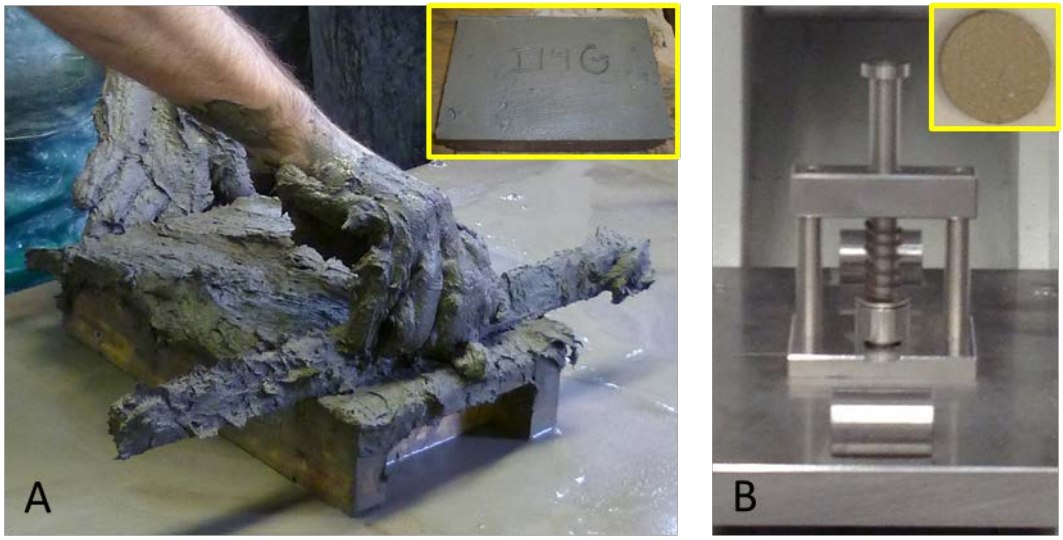

Figure 4. A. The soft-mud method and a moulded sample (green body). B. The dry-pressing pellet mould used in this study and a produced pellet.

Drying and Firing

The drying and firing processes are important in ceramic processing. Cracks and internal stresses can form due to shrinkage, for example in relation to mineral phase transitions (such as the previously mentioned transition in quartz) or decompositions (e.g. liberation of water from clay minerals or carbonate from calcite). At the brickworks Petersen Tegl, where the larger test bricks were fired in a tunnel-kiln (Appendix 3), the heating and cooling rates were lowered in the interval between 520-620 °C in order to avoid a sudden quartz-transition. For the brick-pellet experiments in this study, detrimental internal stresses were less likely to occur irrespective of drying temperature, drying time and heating and cooling rates during firing due to the small size of the pellets. The pellets were therefore fired in a laboratory muffle furnace.

Final products

The produced test-bricks and pellets were characterised according to their appearance, shrinkage, mineralogy, open porosity, bulk density and water absorption. Hygroscopic adsorption, initial freeze-thaw resistance and compression strength were only measured for the test-bricks. Heavy metal leaching was only measured on the waste-containing pellets. A large fraction of the methods employed here investigates aspects of the durability of the brick/pellets. The intrusion of fluids, ice and/or salts in the system can cause the bricks to deteriorate through spalling and cracking. Parameters such as low open porosity, low water absorption and high compression strength are usually good indicators of durability (ASTM C62-13a, 2013; Litvan, 1980; Taylor,

1991). The less porous and impregnable the system is to water, the stronger and more durable it will be. For bricks, some water absorption is, however, required because of the necessary adhesion between mortar and bricks, which requires water to react with the cement (ASTM C216-14, 2014).

Results and discussion

Properties of the marine clay

The investigations from this study (Appendix 1, Appendix 2) found that marine sediments from wide-spread geographical localities in Greenland were very similar, with respect to grain size distribution, mineralogy and major element chemistry. Furthermore it was also established that these properties were very similar to those of North American and Northern Scandinavian marine sediments. In this section some additional data are shown in order to demonstrate the similarities of the sediments. Furthermore, the major element chemistry of these sediments has been compared to brick clays used elsewhere in the World.

Grain size distribution

In Figure 5. the grain size distributions of Greenlandic marine sediments (Appendix 1, Appendix 2 and Foged, 1979) are compared with those of Norwegian and Canadian marine sediments (Foged, 1979; Pederstad and Jørgensen, 1985; Geertsema and Torrance, 2005). A boundary of 63 µm between the silt and sand fraction was used for the grain size distributions on Norwegian sediments reported by Pederstad and Jørgensen (1985). Although this boundary is slightly higher than the 60 µm used in this study, the discrepancy in the ternary plot is believed to be negligible. The grain size distributions of the Canadian marine sediments reported by Geertsema and Torrance (2005) were given as clay, silt and sand fractions, however, the definitions of the fractions were not reported and they are therefore marked as “estimated” in Figure 5. The ranges used in this study: < 2 µm (clay), 2-60/63 µm (silt) and 60/63-2000 µm (sand) are standardly applied and encountered in the literature (DS/EN ISO 14688-1, 2002; Larsen et al., 1995; Pederstad and Jørgensen, 1985) and are therefore also likely to apply to the Canadian sediments. Most of the data points plotted in Figure 5. cluster in the region of ~25-60 wt % clay (< 2µm), 35-70 wt % silt (2-60 µm) and ~0-15 wt % sand (60-2000 µm).

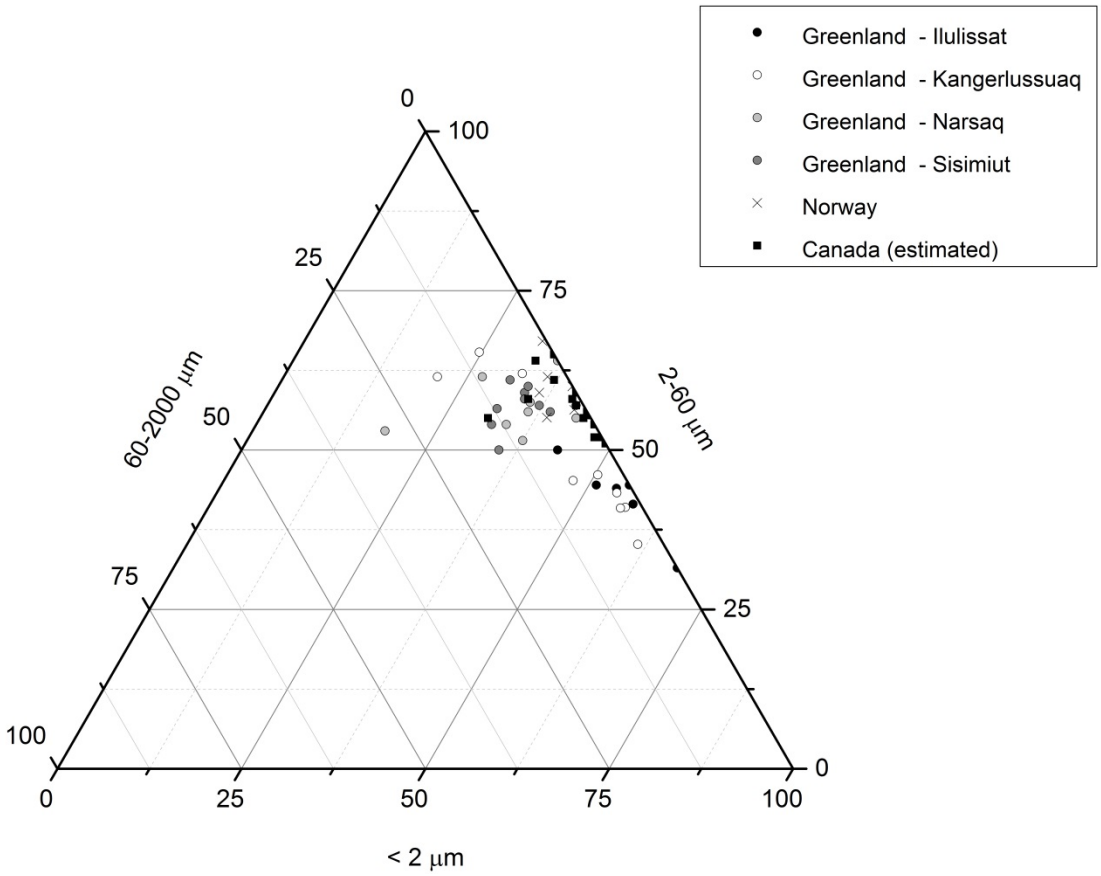

Figure 5. Comparison of grain size distributions of Greenlandic, Norwegian and Canadian marine sediments.

Mineralogy

The Greenlandic, Norwegian and North American marine sediments have a qualitatively similar mineralogy often consisting of feldspars, quartz, amphibole, mica/illite, chlorite, expandable clay minerals (e.g. smectite, vermiculite and mixed layer clays) and calcite and their fine and colloid fractions contain a high percentage of primary minerals such as quartz, feldspars and amphibole (Appendix 1; Foged, 1979; Bentley and Smalley, 1978; Gillott, 1971; Gillott, 1979; Locat et al., 1984; Pederstad and Jørgensen, 1985; Ramesh and d'Angeljan, 1995; Roaldset, 1972).

Major element chemistry

In Table 3 the major element distributions of Greenlandic marine sediments (Appendix 3; Jacobsen et al., 2002 (Appendix 4) and Kalvig, 1990) are compared with those of Norwegian and Canadian marine sediments (Pederstad and Jørgensen, 1985; Roaldset, 1972, Gillott, 1971 and Locat et al., 1984). Furthermore, the marine sediments are compared to brick clays from USA, Europe and China. The Greenlandic sediments generally fall within the ranges of the Canadian sediments,

however, in comparison to the Norwegian sediments the Greenlandic sediments contain slightly more SiO_2 and Na_2O and slightly less Fe_2O_3T (i.e. total Fe_2O_3), MgO and K_2O. This could reflect regional differences in e.g. parent rock composition and would require additional investigations to be confirmed. Overall, the chemical compositions of the marine sediments are dominated by SiO_2 (46-64 wt%), Al_2O_3 (14-23 wt%) and Fe_2O_3T (4-15 wt%) with an alkali-alkaline earth element content (i.e. the content of K_2O, Na_2O, MgO and CaO) of 12-16 wt % (Appendix 3; Bentley and Smalley, 1978; Gillott, 1971; Jacobsen et al., 2002 (Appendix 4); Kalvig, 1990; Locat et al., 1984; Pederstad and Jørgensen, 1985; Roaldset, 1972). The marine sediments compare well with the brick clays in Table 3 although the Na_2O content is higher.

Salt content

The salt content of the marine clay occurrences can vary significantly (Appendix 1), however, most of the uplifted occurrences investigated in this study have low contents of salts due to leaching after uplifting. The Kangerlussuaq occurrence is exceptional, as it appears to be enriched in salts. Although the bricks produced from the Kangerlussuaq occurrence have proved successful, the high salt content could pose a detrimental problem during the firing process, as evaporating salts (i.e. NaCl) can cause corrosion of the kiln.

Plastic properties

The plastic properties of marine sediments from Greenland (Appendix 1; Appendix 2; Foged, 1979; Larsen et al., 2001), Norway (Foged, 1979) and Canada and North America (Locat et al., 1984; Gillott, 1979) are plotted in a Casagrande diagram in Figure 6. The majority of the samples plot above the A-line and classify as clays of low to medium plasticity. The range in plastic properties is, however, quite large, i.e. ~18-70 % for the liquid limit and ~3-45 % for the plasticity index. Locat et al. (1984) found a generally good correlation between the plasticity index and the amounts of phyllosilicates plus amorphous materials of marine clays from Eastern Canada; However, as the amount of neither phyllosilicates nor amorphous material were quantified during this study, it was not possible to confirm such a correlation for the Greenlandic samples.

Locality	Marine sediments					Brick clays		
	Greenland - Ilulissat	Greenland - Kangerlussuaq	Greenland - Qasiannguit	Norway	Canada	Savannah, GA, USA	Italy	China
Reference	Appendix 3	Jacobsen et al. (2002) (Appendix 4)	Kalvig (1990)	Pederstad and Jørgensen (1985); Roaldset (1972)	Gillott (1971); Locat et al. (1984)	Mezencevova et al. (2012)	Dondi et al. (2000)	Chen et al. (2011)
SiO_2 (wt %)	61.1	58.0	64.2	45.8-53.6	46.8-62.6	60.5	42.0-64.6	64.0
TiO_2 (wt %)	0.6	0.8	0.6	0.8-1.1	0.0-1.0	1.0	0.6-1.0	0.9
Al_2O_3 (wt %)	14.5	16.2	14.5	14.3-21.5	15.4-23.4	20.1	10.5-21.2	16.9
Fe_2O_3T (wt %)	6.5	6.6	6.2	8.2-15.2	4.4-8.8	8.1	4.0-7.5	4.4
MnO (wt %)	0.1	0.1	0.1	0.0-0.2	0.1	0.1	0.1-0.2	No data
MgO (wt %)	3.5	3.8	2.4	3.9-5.1	2.1-4.9	1.0	0.8-3.9	1.6
CaO (wt %)	2.4	4.4	3.5	1.0-4.1	2.9-5.4	0.1	0.5-18.0	0.9
Na_2O (wt %)	3.3	4.5	3.4	1.2-2.4	1.3-4.1	0.4	0.5-1.2	0.9
K_2O (wt %)	2.8	2.9	2.7	4.7-6.0	2.4-3.7	2.6	2.1-3.6	2.4
P_2O_5 (wt %)	0.1	0.2	0.1	No data	0.1-0.3	0.1	0.1-0.2	0.2
LOI (wt %)	5.1	2.4	2.4	3.5-10.3	1.6-11.9	6.1	6.0-18.5	7.0

Table 3. Comparison of the major element distribution of marine sediment occurrences from Greenland, Norway and Canada and brick clays from USA, Italy and China.

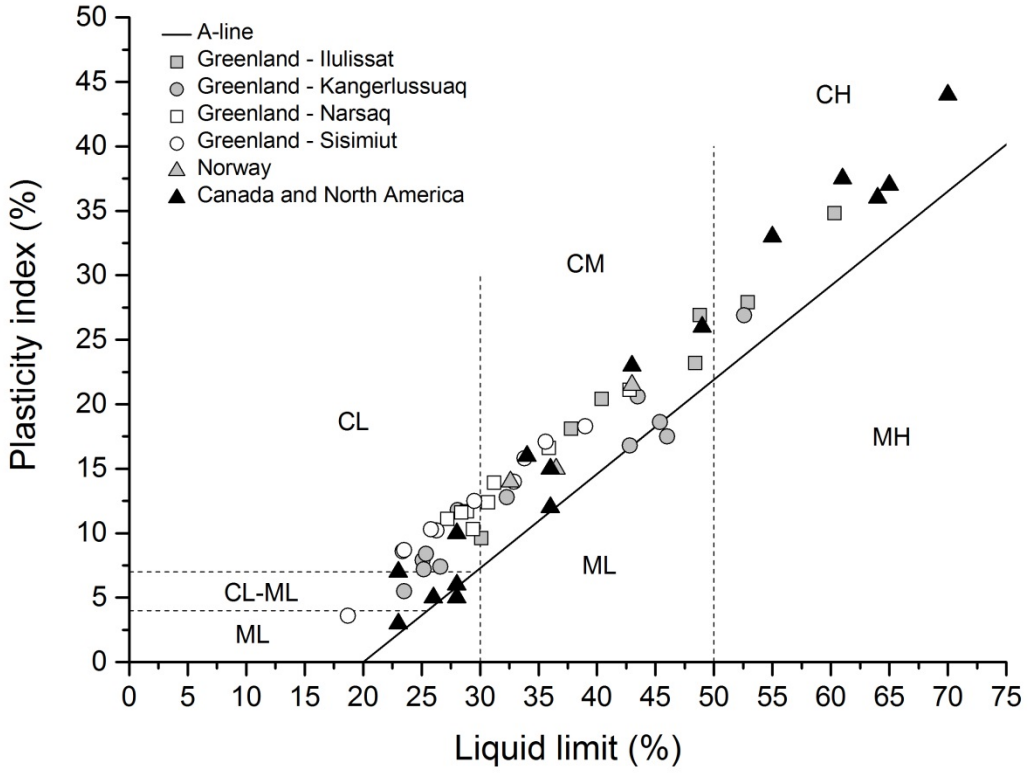

Figure 6. Casagrande plasticity chart (Casagrande, 1948) modified according to Department of the Navy (1986). CL = clay of low plasticity, CM = clay of medium plasticity, CH = clay of high plasticity, ML = silt of low plasticity and MH = silt of high plasticity.

General observations

Despite variations, the Greenlandic, North American and North Scandinavian marine clays have many similarities. This implies that the investigations in respect to brick production will not only be relevant in Greenland, but in a wider geographical area of the arctic to subarctic region. Furthermore, their major element chemistry is comparable to brick clays used elsewhere in the World.

Properties of the test bricks and pellets produced from the marine sediments

Test-bricks and pellets made from pure sediment were investigated first. In Figure 7 the total shrinkage (%), apparent (bulk) density (kg/m^3) and open porosity (%) of the Ilulissat test bricks (Appendix 3) and pellets (Appendix 2 and 6) are plotted against the firing temperature. These

parameters are important in the evaluation of the durability of the produced bricks/pellets. The test bricks have markedly larger total shrinkages and apparent densities and lower open porosities compared to the pellets. These differences are most likely explained by the differences in preparation, production methods (soft mud method versus dry pressing method) and in the drying and firing programmes. In order to directly correlate the results obtained for the pellets with those obtained for the bricks, it would be necessary to use the same production procedure for both.

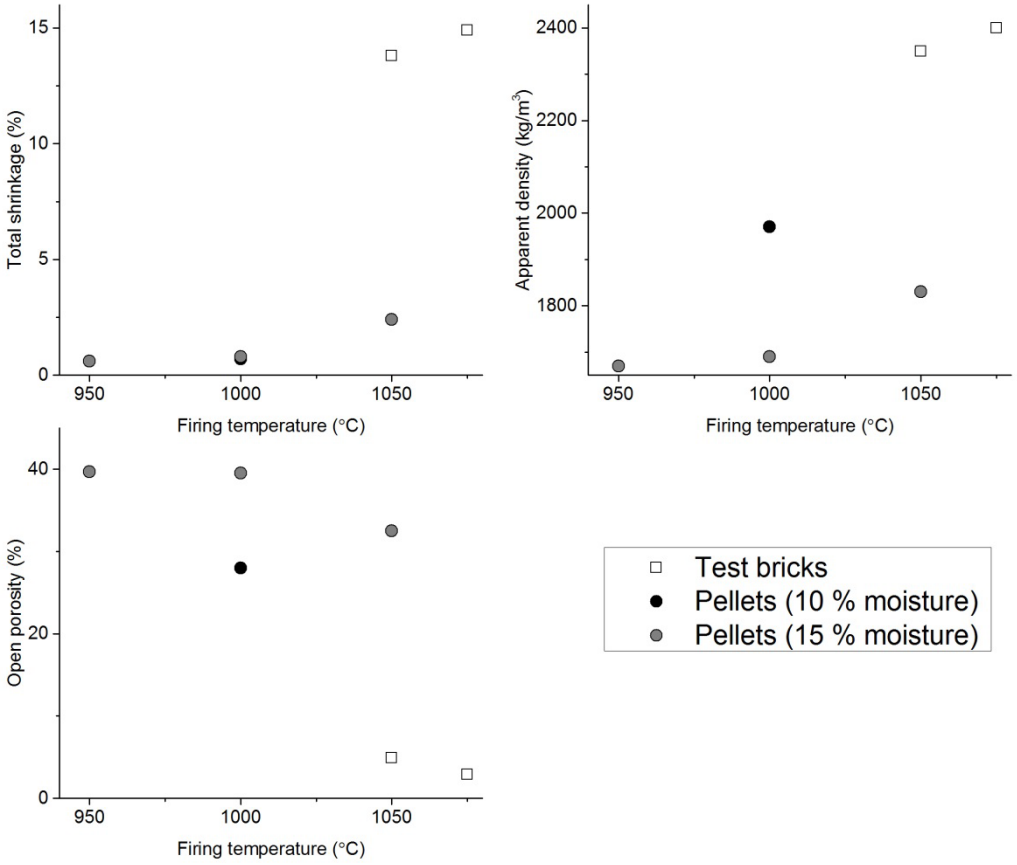

Figure 7. The total shrinkage (%), apparent (bulk) density (kg/m^3) and open porosity (%) of the Ilulissat test bricks and pellets.

Two initial moisture contents (10 % and 15%) were tested for the pellets fired at 1000 °C. The pellets containing 10 % moisture generally had a higher apparent density and lower open porosity than the pellets containing 15 % moisture. The difference is most likely explained by a better initial compaction of the pellets containing 10 % moisture. As the total shrinkages of the two pellet types are very similar, it is unlikely that the pellets containing 10 % moisture should have obtained a

significantly higher degree of sintering. This would, however, have to be confirmed by strength testing on test samples of larger dimensions and SEM imaging before and after firing.

Bulk densities of clay bricks, which are used in the building industry, have been reported to be in the range of 1510-2380 kg/m^3 (Dondi et al., 2004; Dondi et al., 2000; Hall et al., 1992) and open porosities in the range of 18.8-39 vol % (Dondi et al., 2004). The densities (1670-2130 kg/m^3) and open porosities (22-39.7 vol %) found for the pellets (including pellets from Sisimiut and Kangerlussuaq) in this study, are therefore acceptable. For the test bricks, the densities (2350-2400 kg/m^3) are high and porosities (2.9-4.9 %) are low. Furthermore, the test bricks had very high compression strengths (MPa) and low initial rate of water absorption (kg/(m^2·min)) indicating a high degree of sintering. The overall results, however, indicate that acceptable properties can be obtained for Greenlandic marine sediment bricks and that these properties to a large extent can be controlled by preparation, production method and firing programme. Furthermore, this study established an optimal firing temperature in the range from 1000°C-1050°C (Appendix 6).

Properties of the test bricks and pellets produced from a combination of marine sediment and waste materials

After the initial studies on pure sediments, test-bricks and pellets were made using a mixture of the Ilulissat sediment and Greenlandic waste.

Choice of waste materials

Waste materials in Greenland include residential waste, commercial waste, industrial waste from e.g. the fishing and mining industries, and construction and demolition waste (Eisted and Christensen, 2011). The residential and commercial wastes are sorted into combustible and non-combustible fractions. The non-combustible fractions are landfilled and the combustible fractions are incinerated. According to Eisted and Christensen (2011) the industrial waste from the fishing industry consists mainly of organic waste (which is discharged to the sea) and wastes, which are similar to commercial waste (e.g packaging). Larger scale industrial waste (i.e. tailings) from the mining industry is traditionally treated and stored in tailings facilities on-site.

As reported previously the incentive for using waste in bricks is often to either conserve natural resources, to protect the local environment (e.g. by limiting the disposal of hazardous substances) or improve the production costs or quality of the produced bricks (see Table 1). Due to the vast occurrences of Greenlandic marine sediments, the conservation of the natural resources is not an

imminent concern. In Greenland, waste that constitute hazardous substances or has the potential of improving the final product and/or production costs would therefore be of initial interest. In this study, most of the investigated waste materials were therefore of hazardous or potentially hazardous nature. Generally two waste types were selected 1) incineration ashes, e.g. fly ash and bottom ash, from the municipal incineration of residential and commercial wastes and 2) industrial wastes from the mining industry (tailings).

The addition of fly ash generally lowered the apparent (bulk) density and increased the open porosity of the produced pellets compared to the pure sediment pellets, so that values were outside the ranges reported for other construction bricks (Dondi et al., 2004; Dondi et al., 2000; Hall et al., 1992). These properties could however be interesting in lightweight bricks. The addition of bottom ash and tailings, which had a similar or coarser grain size distribution than the sediment, generally gave bulk density and open porosity values within the ranges reported for other construction bricks. Furthermore, lower open porosities and higher densities were generally obtained for the waste/sediment mixtures in comparison to the pure clay composition. This could be an indication that bricks incorporating the bottom ash and tailings could be more durable. Heavy metals appeared to be mobilised after firing for all tested waste/sediment pellet compositions and should be investigated further, as this could pose an environmental hazard.

Another potential waste type, which, however, was not investigated in this study, was construction and demolition waste. Recently, the presence of PCB's has been confirmed in approximately 45 % of the Greenlandic buildings, which were constructed in the period from 1950-1977 (Departementet for miljø og natur, 2014). The demolition wastes from these buildings will therefore likely be classified as hazardous waste. It is estimated that the total amount of PCB's from demolitions will be approximately 200-500 kg and disposal expenses will amount to 200-300 million DKK during the next 10-20 years. As PCB's are destroyed at high temperatures (Qi et al., 2014) it would be interesting to include this type of demolishing waste for future studies on brick production in Greenland.

Concerns regarding the use of waste materials in Greenlandic brick production

The modern brick industry is highly dependent on technology and requires high capital investments. In order to ensure an optimal production and yield, it is therefore important to obtain consistent raw materials with predictable firing characteristics (Bloodworth et al., 2007). This presents a dilemma, when new raw materials, such as MSWI ashes and mine tailings are

concerned. These materials are often heterogeneous and would either require large scale homogenisation or treatment, such as fractionation into more homogeneous fractions. The composition of MSWI ashes could be controlled by limiting the types of waste that were incinerated in a specific plant. A tendency towards this is emerging worldwide. In Denmark, for example, the recycling of waste is planned to increase significantly and will ultimately change the composition of the waste that is finally incinerated (the Danish Government, 2013). However, due to the small size of the Greenlandic population and it's wide geographical distribution it is questionable whether a similar approach will be practical in Greenland (Eisted and Christensen, 2013). Kalvig (1990) suggested that a small-scale mobile brick plant could be attractive in Greenland, due to low capital investments and minor requirements for labourers. These plants are designed to be moved from place to place and are perhaps better to suited to incorporate wastes of a more heterogeneous nature. Another approach could be to dilute the waste, i.e. only incorporate small amounts of waste into each brick production, and thereby lower the environmental hazards.

Conclusions

Large marine sediment resources are common in Greenland and are often in close vicinity to towns and settlements. In this phd study their similarities in respect to grain size distribution, mineralogy and major element chemistry was established. Differences in salt content and plastic properties will influence the suitability or production method for a specific sediment occurrence. The Greenlandic marine sediments are similar to marine clay occurrences in Northern America and Northern Scandinavia, which indicate that the conclusions from this project is not only relevant in Greenland but also in the wider Arctic to subarctic area. The test bricks and pellets produced from Greenlandic marine sediments generally obtained acceptable properties of porosity and density. However, these properties were shown to vary significantly and be highly dependent on the processing, e.g. firing programme and initial treatment. This project therefore concludes that it is possible to produce good quality bricks from the marine sediments. The next step should be an optimisation of the raw material composition and processing in order to obtain bricks, which will be suited to the Arctic to sub-arctic climate.

The bottom ash and tailings, which were investigated in this study, can be included without harming or even enhancing technical properties such as porosity and density. The fly ashes generally reduced density and increased porosity, which could be an interesting property for e.g.

lightweight bricks. Leaching tests, however, indicate that some heavy metals become more mobile after firing and further and larger scale testing will therefore be needed in the future.

Overall, the technical investigations from this project have shown that it is possible to produce bricks of good quality from the vast local resources of marine sediments in Greenland.

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Appendix 1

Description: Unpublished paper

Title: Characterisation and weathering properties of fine-grained marine sediments from West Greenland

Characterisation and weathering properties of fine-grained marine sediments from West Greenland

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Abstract

The understanding of fine-grained marine glaciogene sediments, which are found throughout the former glaciated part of the northern hemisphere, is of immense importance in geotechnical and civil engineering. In Greenland, however, the properties of these sediments have not been studied to any great extent, in comparison to marine sediments from Canada and Northern Scandinavia. In this study, samples from boreholes in four towns in West Greenland have been characterised according to their grain size, mineralogy, Atterberg limits and pore water chemistry. The samples were selected in order to represent environments above and below present day sea level, as well as thermal conditions with and without permafrost. On the basis of the pore water chemistry we were able to identify three types of profiles: marine, enriched and depleted. Furthermore, the analyses indicate that the Greenlandic sediments are very similar to the sediments from Canada and Northern Scandinavia and classify as clays of low to high plasticity. The content of expandable clay minerals in the clay fraction indicate that chemical weathering has been an active process during glacial erosion of bedrock, in deposition in seawater and/or after subaerial exposure.

Keywords: Greenland; marine clay; weathering; clay mineralogy; standard classification tests; pore water chemistry.

1. Introduction

Deposits of fine-grained marine glaciogene sediments have been observed throughout the former glaciated parts of the Northern hemisphere. They were formed by glacial erosion and deposited in marine environments by the end of the last glaciation. Due to a generally high sensitivity and low remoulded strength, these sediments have been known to cause severe subsidence and disastrous landslides (e.g. the Saint-Jean-Vianney slide in Canada (Tavenas et al., 1971; Smalley et al., 1975), the Rissa slide in Norway (Gregersen, 1981)) and have therefore been of special interest to geotechnical engineers.

In Northern Scandinavia, Canada and Alaska much scientific attention has been given to the characterisation of these sediments (Gillott, 1971; Roaldset, 1972; Bentley and Smalley, 1978; Gillott, 1979; Locat et al., 1984; Pederstad and Jørgensen, 1985; Ramesh and d'Angeljan, 1995). The sediments are generally fine-grained with the < 2 μm fraction constituting 20-94 wt % and they have a qualitatively similar mineralogy often consisting of feldspars, quartz, amphibole, biotite/illite, chlorite, expandable clay minerals (e.g. smectite and vermiculite) and calcite. Their fine and colloid fractions contain a high percentage of primary minerals (such as quartz, feldspars and amphibole) and the proportion of expandable clay minerals can vary considerably from region to region. Furthermore, their plastic properties mostly classify them as clays ranging from low to high plasticity according to Casagrande (1948).

In Greenland, much less scientific attention has been given to the characterisation of these occurrences and soils and glaciogene sediments have in general been described as rock flour (Jensen, 1965; Jensen et. al., 1974; Heling, 1976), based on an assumption that chemical weathering is negligible in the arctic climate. Plasticity measurements in order to support this description are, however, very scarce and mineralogical investigations on the < 2 μm fraction conducted by Jensen (1965) and Heling (1976) seem to indicate that the Greenlandic sediments

are similar to the clay deposits of Northern America and Scandinavia. With the continuing deposition of glaciogene sediments into the marine environment, due to the presence of the Ice sheet, Greenland, nevertheless, provides a unique opportunity to study the formation and development of these sediments.

The area around the town of Kangerlussuaq represents such a unique and very young geological environment, where glaciogene sediments are still deposited in a marine fjord environment today. Wimpenny et al. (2010) studied the glacial effects on weathering processes in this area and estimated chemical and physical erosion rates of 10 t/km^2/yr and 1560 t/km^2/yr, respectively. Although these figures indicate that mechanical weathering is predominant in the glacial erosion in Kangerlussuaq, chemical weathering is still an active and significant process. West et al. (2005) have reported similar chemical erosion rates from alpine catchments with partial glacial cover in North America and Scandinavia. Furthermore, the straining and abrasion in connection with glacial erosion might make minerals more susceptible to chemical weathering (e.g. Newmann and Brown, 1969) during transportation and deposition, which is consistent with observations of mild chemical weathering after deposition on Canadian and Norwegian marine glaciogene sediments (Ramesh and d'Anglejan, 1995; Pederstad and Jørgensen, 1985).

In this study, we have investigated marine glaciogene sediments from Kangerlussuaq and combined our data with previous investigations on marine sediments from Ilulissat, Sisimiut and Narsaq (Foged 1975, 1979). The combined dataset presented here represents marine glaciogene sediments of both currently marine and subaerially exposed environments. Furthermore, samples and locations have been selected to represent thermal conditions with and without permafrost. This provides a unique dataset for discussing the formation and describing the development of the fine-grained marine sediments in western Greenland with respect to chemical weathering.

2. Materials and methods

2.1 Geological setting

At the last glacial maximum approximately 25 ka BP, all of West Greenland was covered by the Greenland Ice Sheet, which is believed to have extended 100 km beyond the present coast line (Weidick, 1976; van Tatenhove et al., 1996). The retreat of the Greenland Ice sheet following the last glacial maximum and the resulting marine transgression resulted in the deposition of fine-grained marine sediments in large parts of West Greenland, due to flocculation of glacial river sediments upon entering the saline marine environment. Isostatic rebound has since elevated some of these sediments above present day sea level, exposing them to percolating precipitation and thus depletion of the saline pore water. The general climatic cooling in West Greenland which marked the end of the Holocene optimum around 5 ka BP (Dahl-Jensen et al., 1998) induced the formation of ice-rich permafrost in some of these sediments, effectively stopping the depletion process.

Regional differences in the isostatic uplift have resulted in differences in the timing and duration of exposure to depletion, and thus the fine-grained deposits found in some regions may be fully (or nearly) leached, while those in other regions may be only partly leached. These differences strongly affect the freezing temperature of the sediments and the presence and distribution of ice features in permafrost regions, but may also affect the possibility for chemical weathering.

In this study we selected fine-grained sediment samples of glaciomarine origin from boreholes in four towns in West Greenland: Kangerlussuaq, Ilulissat, Sisimiut and Narsaq (see Figure 1). The borehole and sample details are given in Table 1 and maps indicating the borehole locations are presented in Figure 2.

The **Kangerlussuaq** area was deglaciated approx. 8 ka BP (van Tatenhove et al., 1995; Bennike and Björk, 2002; Roberts et al., 2009) and was exposed by marine regression after 5.9 ka BP, which was demonstrated by recent C^{14} dating of marine shells (Holtegaard Nielsen, pers. comm.). The samples analysed in this study were collected from two boreholes in different settings. Borehole KAN A is situated 3 km South West of the airport on an elevated marine terrace, *Fossilsletten* (18.5 m a.s.l.), in direct hydrological contact with the saline fjord waters. Arid conditions in the area have resulted in formation of dry-crust and concentration of ions in the upper parts of the profile due to capillary rise. Borehole KAN B is located 4 km to the east of the airport (away from the fjord) in a bedrock confined basin (34.3 m a.s.l.). Although Kangerlussuaq is located in the continuous permafrost zone and the mean annual ground temperature (MAGT) has been observed to be -1.6 °C (at 4 m depth; Christiansen et al., 2010), the high salinity has prevented ground ice formation at borehole KAN A, whereas the sediments encountered in borehole KAN B are extremely ice-rich and highly sensitive (Agergaard and Ingeman-Nielsen, 2010).

The **Ilulissat town area** was deglaciated approximately 9.5 ka BP (Weidick and Bennike, 2007), and probably slightly later than the adjacent coastal areas of the inner Disko Bay (Long and Roberts, 2002), due to the vicinity to the major outlet glacier, Kangia. Samples were selected from the two boreholes ILU A and ILU B. The latter was drilled in the sea bed of the harbour basin, *Kanelen*, at elevation -5.0 m a.s.l., as part of the site investigations for harbour piers. Studies of the upper marine limit from surrounding areas indicate that the sediments in the borehole have been subjected to marine conditions since their deposition. Borehole ILU A is located in a bedrock confined basin at +53.3 m a.s.l. This area is believed to have been exposed by marine regression 6.5-7 ka BP (Rasch, 2000). Ilulissat is located in the continuous permafrost zone, with MAGT around -3.4 °C (at 4 m depth; Christiansen et al., 2010), and the fine-grained sediments encountered in ILU A were frozen with occasional thin ice lenses.

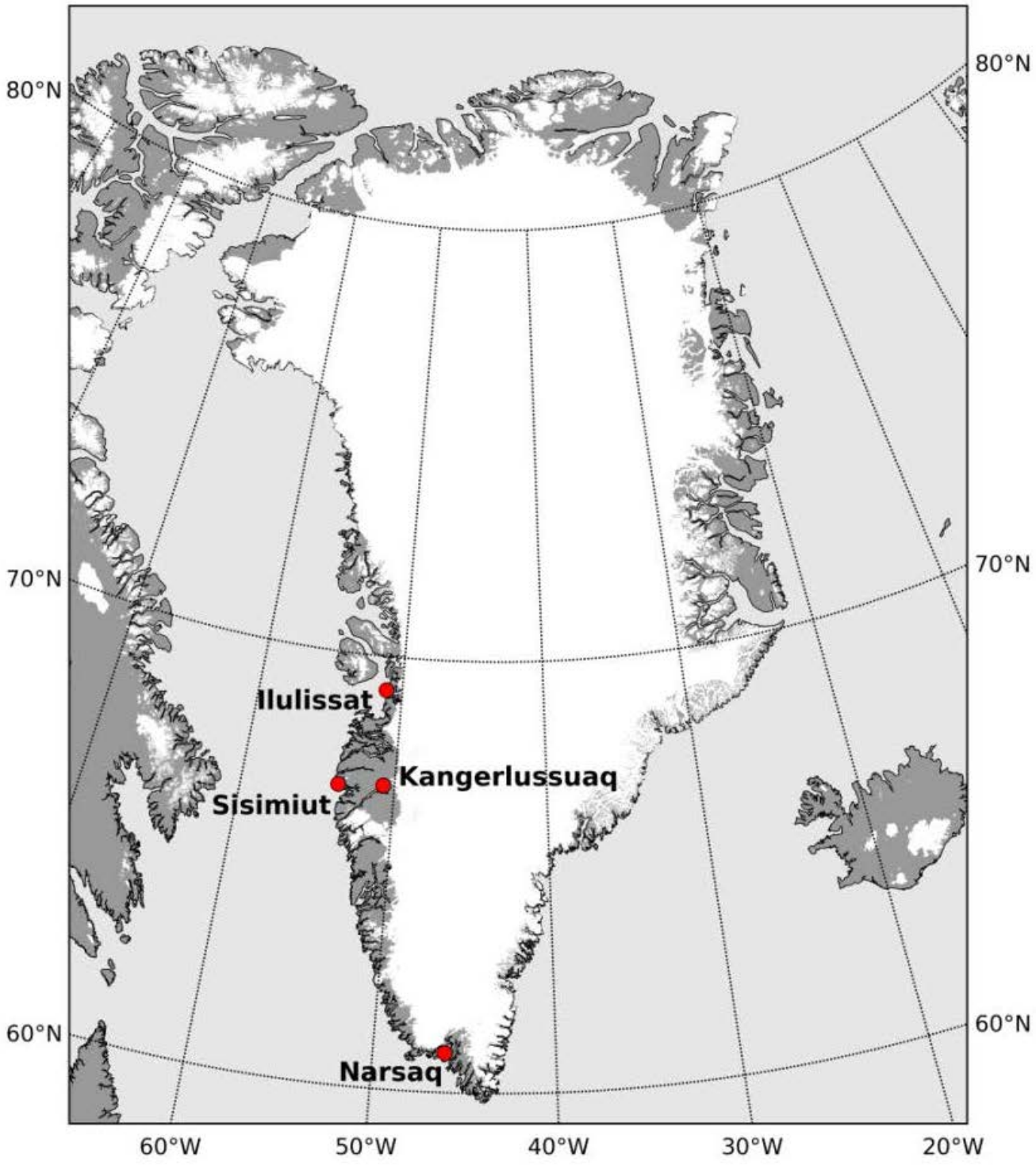

Figure 1. Map of Greenland with the four towns indicated.

Locality	Borehole no. (and ID)	Coordinates	Elevation (m a.s.l.)	Sample name	Depth (m b.s.)	Original sample name
Kangerlussuaq, Fossilsletten	KAN2005-04 (KAN A)	E509882 N7431711	+18.5	KAN A-1	0.0	No. 1
				KAN A-2	1.0	No. 4
				KAN A-3	1.8	No. 7
				KAN A-4	3.0	No. 9
				KAN A-5	3.8	No. 12
				KAN A-6	5.0	No. 14
Kangerlussuaq, River bank	KAN2009-01A (KAN B)	E517560 N7433759	+34.3	KAN B-1	4.2	Kang-gr1A-6E
				KAN B-2	4.6	Kang-gr1A-7E
				KAN B-3	5.4	Kang-gr1A-10A2
Ilulissat, Town	GTO72034 (ILU A)	E496316 N7678020	+53.3	ILU A-1	1.8	JAK 38
				ILU A-2	3.8	JAK 40
				ILU A-3	5.4	JAK 41-2
				ILU A-4	7.3	JAK 43-1
Ilulissat, Kanelen	GTO71137 (ILU B)	E496285 N7678962	-5.0	ILU B-1	3.9	JAK 253
				ILU B-2	5.9	JAK 257
Sisimiut, Parassuq	GTO74063 (SIS A)	E382607 N7426989	-2.2	SIS A-1	0.4	HBG 181
				SIS A-2	2.5	HBG 185
				SIS A-3	4.5	HBG 189
				SIS A-4	6.5	HBG 193
				SIS A-5	8.6	HBG 197
Sisimiut, Pamap Kua	GTO69011 (SIS B)	E384334 N7427037	+48.8	SiS B-1	1.2	HBG 368
				SIS B-2	1.9	HBG 370
				SIS B-3	3.5	HBG 373
				SIS B-4	3.8	HBG 376
Narsaq, Centre	GTO74017 (NRQ A)	E443296 N6753655	+24.2	NRQ A-1	2.3	NRQ 287
				NRQ A-2	2.8	NRQ 288
				NRQ A-3	4.3	NRQ 290
				NRQ A-4	5.5	NRQ 292
				NRQ A-5	6.8	NRQ 293
				NRQ A-6	8.4	NRQ 295
Narsaq	GTO74012 (NRQ B)	E443247 N6754208	+ 14.7	NRQ B-1	1.1	NRQ 20
				NRQ B-2	2.0	NRQ 21

Table 1. Borehole and sample information. Coordinates are given in UTM WGS84 zone 22N for all boreholes except the boreholes from Narsaq, which are zone 23N. Elevations are given as meters above mean sea level (m a.s.l.) and depths are given as meters below ground surface or sea bed (m b.s.). In this paper, a simplified borehole ID and sample name are applied in order to provide clarity. Original borehole no. and sample names are included in the table for reference.

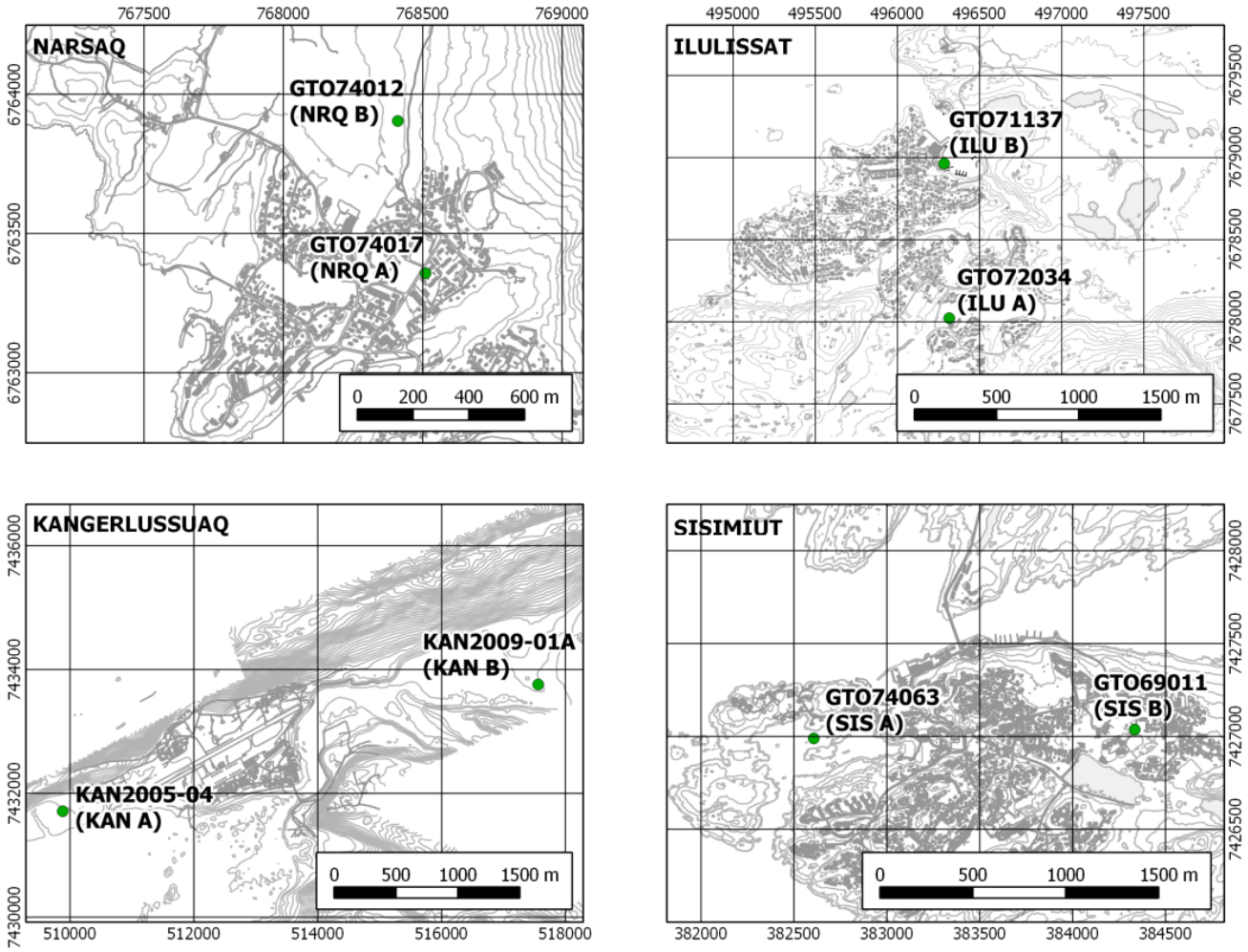

Figure 2. Maps of the four towns Narsaq, Kangerlussuaq, Ilulissat and Sisimiut with indication of borehole locations. Topographic equidistance is 10 meters in all maps. Coordinates are given in UTM WGS84 zone 22N for all maps except the map of Narsaq which is zone 23N. Please also note that the four maps are rendered at different scales.

In the **Sisimiut area**, samples were collected from boreholes SIS B and SIS A in two bedrock confined sedimentary basins: *Pamap Kua* at 48.8 m a.s.l. and *Parassuq* around present day sea level (sea floor at -2.2 m a.s.l.) The Sisimiut area was deglaciated approx. 10 ka BP (van Tatenhove et al., 1996; Bennike and Björk, 2002; Roberts et al., 2009) and the Pamap Kua basin was exposed by the marine regression approximately 9 ka BP (Bennike et al., 2011). A borehole adjacent to SIS B shows development of dry crust. Sisimiut is located in the discontinuous permafrost zone, and at the time of drilling, ice lenses were observed in the major part of the fine-grained sequence (down to the boundary with coarse grained sediments at 6 m b.s.) The borehole SIS A is situated below

present day sea level, and observations do not demonstrate the development of dry crust (preconsolidation by water removal), indicating that the sediments have not been extensively exposed to freeze/thaw processes or subaerial conditions. This is consistent with reports of a very limited, late Holocene sea level rise in the area (Bennike et al., 2011; Long et al., 2010; Roberts et al., 2009). However, locally in another part of this basin a zone with high overconsolidation was found, most likely caused by subaerial exposure and drying of these sediments.

Samples from the **Narsaq area** were collected from boreholes in two bedrock confined sedimentary basins: Borehole NRQ A is located 24.2 m above mean sea level (m a.s.l.), while borehole NRQ B is located 14.2 m a.s.l. The Narsaq area was deglaciated prior to 10.6 ka BP (Bennike and Björk, 2002), and the marine regression exposed the basins around 10 ka BP (Bennike et al., 2002; Sparrenbom et al., 2006). The marine regression continued in the area for several millennia to a relative sea level approximately 10 m lower than present day (Kuijpers et al., 1999; Sparrenbom at al., 2006). Today, the areas are not affected by permafrost, and no indications of previous permafrost conditions have been observed. However, the sedimentary sequences observed from the boreholes are strongly affected by dry crust probably due to evaporation and/or the annual winter freeze/thaw processes. The dry crusts are underlain by clays of high sensitivity in both boreholes.

2.2 Methods

The analysis of samples from the two Kangerlussuaq boreholes were made specifically for this study, and this section presents the details of the methodology used for the determination of particle size distributions, Atterberg limits, mineralogy and chemistry of the KAN A and KAN B samples. The remaining analyses were performed by Foged (1975; 1978; 1979) as part of his PhD study and a review of his methodology is presented in appendix A. In this study we have chosen to apply methods, which are identical or directly comparable to the methods used by Foged.

2.2.1. Particle size distributions

The particle size distributions were analysed by a combination of sieving (using wet preparation) and sedimentation (using the hydrometer method) in accordance with DS/CEN ISO/TS 17892-4:2004. In this study we have not analysed the detailed distribution of the < 2 µm fraction.

2.2.2. Atterberg limits and loss on ignition

The natural water content (w_{nat}) was measured by drying the samples at 105-110°C for more than 14 hours, and determined as the percentage mass loss compared to dry mass. This procedure follows the principles of DS/CEN ISO/TS 17892-1:2004.

The liquid limits (w_L) of the KAN A samples were determined by use of the Casagrande method in accordance with BS 1377:1975 and ASTM D423-66:1972. The liquid limits (w_L) of the KAN B samples were measured using the fall-cone method in accordance with DS/CEN ISO/TS 17892-12:2004. Plastic limits (w_P) were determined by the rolling method in accordance with DS/CEN ISO/TS 17892-12:2004.

The loss on ignition (LOI) of the KAN B samples was determined as the weight loss between 110°C and 600°C and was measured after heating at 600°C for 4 hours.

2.2.3 Mineralogy

2.2.3.1 Sample preparation and measurement

The samples were dispersed in distilled water and the < 2 µm fraction was separated by sedimentation. The < 0.2 µm was separated by centrifuge, using the integrated form of Stoke's law for calculations of centrifugation times (Jackson, 1985), and flocculated by use of approximately 10 ml 1 M $MgCl_2$ per 100 g of suspension. Both fractions (0.2-2 µm and < 0.2 µm) were subsequently saturated with $MgCl_2$, according to the method described by Moore and Reynolds (1997) and

orientated samples were prepared by use of the smear mount method (see e.g. Moore and Reynolds, 1997). The orientated samples were subjected to treatments of air-drying, ethylene glycolation (vapour) at room temperature for at least 48 hours and heating at 350°C and 550°C, respectively, for a minimum of 2 hours. The samples were analysed by X-ray diffraction (XRD) on a PanAlytical X'Pert Pro, equipped with Ni-filter and X'Celerator detector. Cu K_α radiation was generated using 45 kV and 40 mA, and a scan speed of 0.008 °2Θ/s was used for data collection.

The octahedral occupation of illite was determined by use of the (060)-reflection (Brown and Brindley, 1980) measured on unorientated powder samples prepared for selected samples. The measurements were carried out on a PanAlytical X'Pert Pro diffractometer, equipped with Ni-filter and using Cu K_α radiation generated at 45 kV and 40 mA.

2.2.3.2. Mineral identification and semi-quantitative distribution

The mineral phases were identified according to their behaviour after treatments of air-drying, ethylene glycolation and heating at 350°C and 550°C, respectively (Brown and Brindley, 1980). The phases studied were smectite, vermiculite, illite, mixed layer clay minerals (regular) and unidentified expandable clay minerals (random mixed layer), chlorite and kaolinite. The name illite is applied here as a general term of all micas in the < 2 μm fraction in accordance with the original definition given by Grim et al. (1937). Other mineral phases, such as quartz, feldspar and amphibole, were observed in all sample fractions but were not included in the semi-quantitative calculations.

The semi-quantitative weighted peak area ratio (WPAR) method used by Foged (1975, 1978, 1979) and in this study is similar to the methods used by Biscaye (1965) and Johns et al. (1954). The weighting scheme used for the Greenlandic samples was developed by Foged (1978) and is listed in Table 2. In this study, peak area intensities were determined as the peak height above

background times the FWHM (full width at half maximum), a procedure which has previously been described by Norrish and Taylor (1962). The equations for the calculation of the WPAR for each of the mineral components are given in Table 2.

2.2.4. Chemistry

4 g of dried material from each sample was dispersed in 50 ml distilled H_2O and filtrated through a 0.45 µm filter by use of vacuum pumping. Cl^- was analysed by ion chromatography (IC). Mg^{2+}, Ca^{2+}, Na^+ and K^+ were analysed by atomic absorption spectroscopy (AAS) for the KAN A samples and by inductively coupled plasma optical emission spectrometry (ICP-OES) for the KAN B samples. Results were corrected for initial content of pore water, i.e. natural water content.

3. Results

The grain size distributions are presented in Figure 3 (A). All samples are dominated by the < 60 µm fraction (clay and silt fractions) and contain only minor amount of the 60-200 µm fraction (sand fraction). Most samples from Kangerlussuaq (KAN A) and Ilulissat (ILU A and ILU B) contain around or more than 50% grains in the <2 µm fraction (clay fraction), whereas the remaining samples range mainly from 20 to 40% in the <2 µm fraction.

	Approximate peak position (d-value)	Intensity weighing factor (W)	WPAR calculation
Total intensity (T)	-	-	$I_T = W_{10Å} \cdot \max(I_{10Å\,(350)}, I_{10\,Å\,(550)}) * + W_{7Å} \cdot I_{7Å\,(350)}$
Smectite (S)	17 Å	$W_{17Å}$ = 1:4	$I_S = \frac{W_{17Å} \cdot I_{17Å\,(glyc)}}{I_T}$
Vermiculite (V)	14 Å	$W_{14Å}$ = 1:2,5	$I_V = \frac{W_{14Å} \cdot (I_{14Å\,(glyc)} - I_{14Å\,(350)})}{I_T}$
Mixed layer clays (ML)	12 Å	$W_{12Å}$ = 1:1	$I_{ML} = \frac{W_{12Å} \cdot I_{12Å\,(350)}}{I_T}$
Unidentified expandable clay (UE)	10 Å	$W_{10Å}$ = 1:1	$I_{UE} = \frac{W_{10Å} \cdot \max(I_{10Å\,(350)}, I_{10\,Å\,(550)}) *}{I_T} - I_{IL} - I_S - I_V - I_{ML}$
Illite (IL)	10 Å	$W_{10Å}$ = 1:1	$I_{IL} = \frac{W_{10Å} \cdot I_{10Å\,(glyc)}}{I_T}$
Chlorite** (C)	7 Å	$W_{7Å}$ = 1:2	$I_C = \frac{W_{7Å} \cdot I_{7Å\,(350)}}{I_T}$

Table 2. Approximate positions of peaks used for the WPAR calculations, Intensity weighing factors and WPAR calculations for the mineral phases studied here. The individual weighing factors (W) and formulas for the WPAR calculations were developed by Foged (1978). The peak area intensity (I) is the area of a given peak corrected for background and overlap from other peaks. *In some diffractograms, an increase in the area of the 10Å peak at 550°C compared to the 350°C peak was observed. This was interpreted as expandable mineral phases that required a higher temperature than 350 °C in order to collapse to 10Å. In these cases the weighted peak area at 550°C was used to calculate the I_T and the I_{UE}. ** Chlorite was distinguished from kaolinite based on the (001) reflection at 14 Å for samples heated at 550°C. Furthermore, the kaolinite (002) and chlorite (004) reflections at 3,57 and 3,53 Å, respectively, were used for identification. No distinguishable kaolinite was observed in the Greenlandic samples.

The mineralogical WPAR distributions of the 0.2-2 µm and < 0.2 µm grain size fractions are presented in Figure 3 (B and C, respectively). With few exceptions, all samples show an increase in the total content of expandable minerals (i.e. smectite, vermiculite, unidentified expandables and mixed layer clay) in the < 0.2 µm fraction compared to the 0.2-2 µm fraction, with a corresponding decrease in the illite and chlorite content. As an example, X-ray diffractograms of the 0.2-2 µm and < 0.2 µm fractions for sample KAN A-3 are shown in Figure 4. The increased WPAR fraction of expandable minerals in the < 0.2 µm fraction is evident from the increased combined peak areas of the broad glycolated ~16-17 Å and ~14 Å peaks compared to the glycolated peaks in the 0.2-2 µm fraction. Similarly, the reduced fraction of the illite peak at ~ 10 Å is also evident.

The illites were confirmed to be dominantly trioctahedral (biotite-type) in all the analysed samples, based on clear peaks at 1.54 Å and very weak (or non-existing) peaks at 1.50 Å (not shown).

The results of the standard classification tests are presented in Table 3. The samples KAN B (all), SIS A (2,3,4,5), SIS B (1,3) and NRQ A (3,4,5,6) have natural water contents (w_{nat}) higher than the liquid limit (w_L), causing the sediments to be highly sensitive to quick. This property has most likely resulted from leaching of salts from the pore water as described by Rosenqvist (1966). The LOI is usually taken as a measure of the organic content in a sample, however, due to the presence of clay minerals, the measured LOI will also include contributions from liberation of crystal water during initial decomposition of these phases. The LOI can, therefore, be considered to be a maximum value of organic content in the samples. For the samples studied here, the LOI is below 2.5 %, with an average of 1.3 %, and indicates a low organic content, which is consistent with the inorganic origin of the sediments. The activities range from 0.25-0.78 and the samples are generally classified as inactive to normal according to Skempton (1953), indicating variations in the weathering of the different samples.

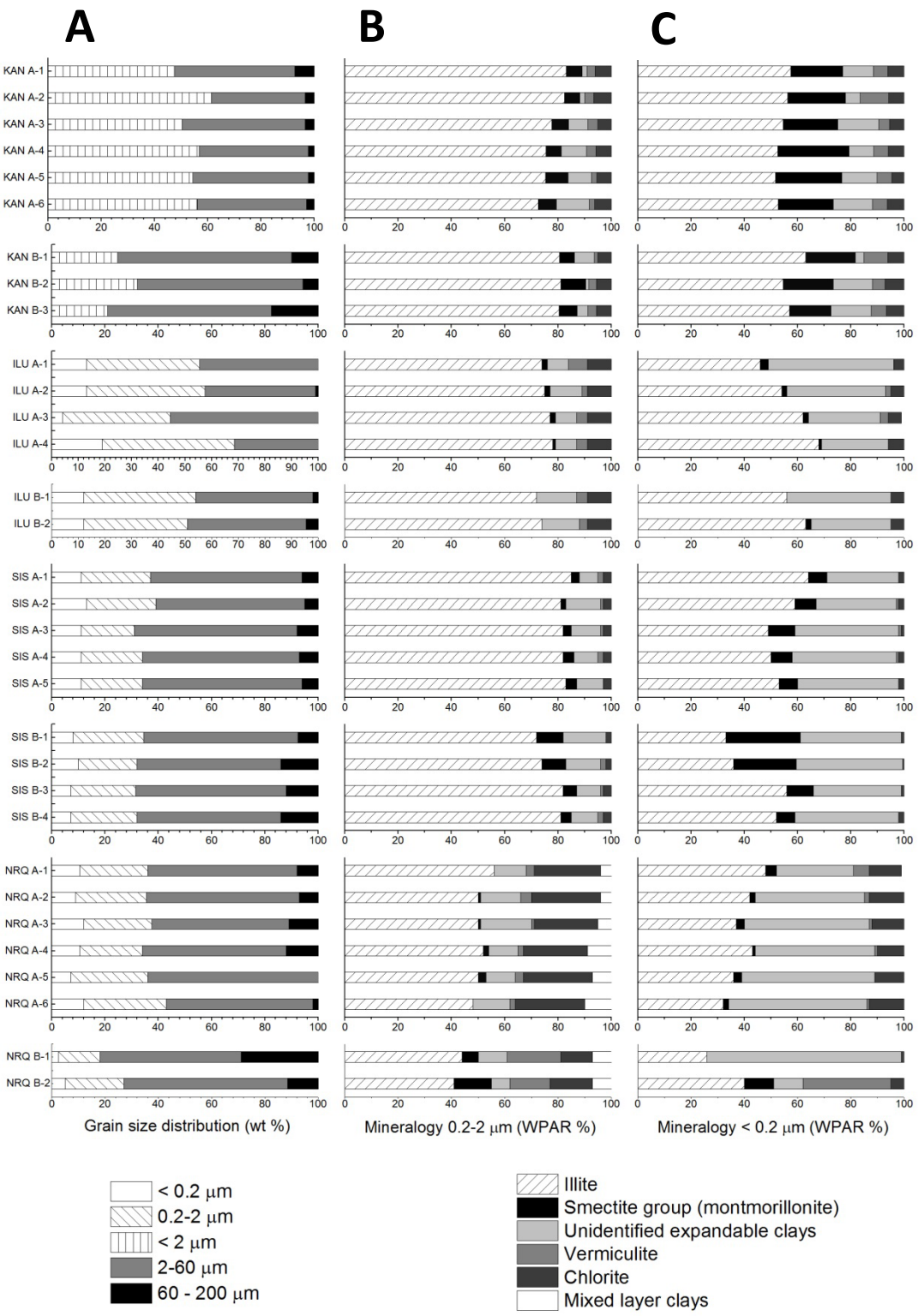

Figure 3. Grain size distribution (A) and WPAR mineralogy of the 0.2-2 μm (B) and < 0.2 μm (C) fractions.

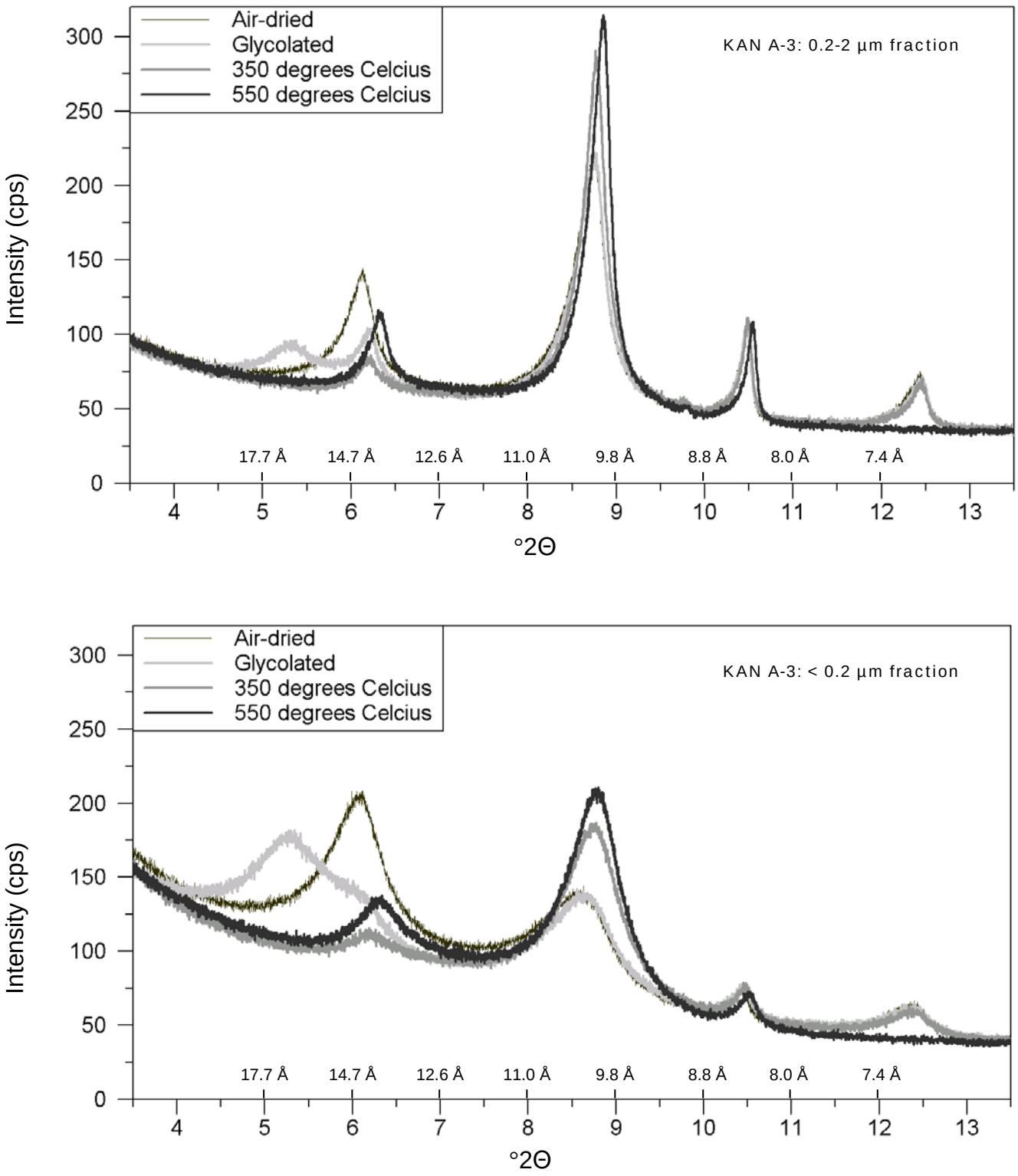

Figure 4. X-ray diffractograms of the 0.2-2 µm and < 0.2 µm fractions of the KAN A-3 sample after treatments of air-drying, glycolation and heating at 350 °C and 550°C, respectively. Approximate d-values are indicated above the °2Θ values for clarity. The peak at ~10.5 °2Θ (8,4 Å) was assigned to amphibole.

Sample Name	Depth (m.b.s.)	LOI (%)	w_{nat} (%)	w_P (%)	w_L (%)	PI (%)	A
KAN A-1	0.0	N.D.	20	16.3	28.1	11.8	0.25
KAN A-2	1.0	N.D.	32	25.9	42.8	16.8	0.28
KAN A-3	1.8	N.D.	31	26.8	45.4	18.6	0.37
KAN A-4	3.0	N.D.	40	28.5	46.0	17.5	0.31
KAN A-5	3.8	N.D.	47	25.7	52.6	26.9	0.55
KAN A-6	5.0	N.D.	36	23.0	43.5	20.6	0.37
KAN B-1	4.2	0.5	31	17.2*	25.1*	7.9*	0.32
KAN B-2	4.6	1.0	33	17.0*	25.4*	8.4*	0.26
KAN B-3	5.4	0.6	27	18.0*	25.2*	7.2*	0.34
ILU A-1	1.8	1.5	40	25.2	48.4	23.2	0.42
ILU A-2	3.8	1.6	37	25.0	52.9	27.9	0.49
ILU A-3	5.4	1.4	36	19.7	37.8	18.1	0.41
ILU A-4	7.3	1.9	41	25.5	60.3	34.8	0.51
ILU B-1	3.9	2.0	37	21.9	48.8	26.9	0.50
ILU B-2	5.9	2.4	37	20.0	40.4	20.4	0.40
SIS A-1	0.4	0.9	29	18.9	32.9	14.0	0.38
SIS A-2	2.5	1.2	41	20.7	39.0	18.3	0.47
SIS A-3	4.5	1.1	33	17.0	29.5	12.5	0.40
SIS A-4	6.5	1.1	40	18.0	33.8	15.8	0.46
SIS A-5	8.6	1.3	40	18.5	35.6	17.1	0.50
SiS B-1	1.2	0.8	26	14.8*	23.4*	8.6*	0.25
SIS B-2	1.9	0.7	22	14.8*	23.5*	8.7*	0.27
SIS B-3	3.5	0.6	28	16.1	26.3	10.2	0.32
SIS B-4	3.8	0.6	25	15.5*	25.8*	10.3*	0.32
NRQ A-1	2.3	1.1	23	17.2	28.9	11.7	0.33
NRQ A-2	2.8	1.6	25	17.3	31.2	13.9	0.39
NRQ A-3	4.3	1.2	31	16.8	28.4	11.6	0.31
NRQ A-4	5.5	1.5	35	16.1	27.2	11.1	0.33
NRQ A-5	6.8	1.4	40	18.3	30.7	12.4	0.34
NRQ A-6	8.4	1.9	38	19.3	35.9	16.6	0.39
NRQ B-1	1.1	1.5	20	19.1	29.4	10.3	0.57
NRQ B-2	2.0	1.4	29	21.7	42.8	21.1	0.78

Table 3. Standard classification tests: loss on ignition (LOI), natural water content (w_{nat}), plastic limit (w_P) and liquid limit (w_L). The plasticity index (PI) is calculated as w_L-w_p. The activity (A) is calculated as PI divided by the percent of the < 2 μm fraction in the total sample (Skempton, 1953). * indicate that values were measured on adjacent samples.

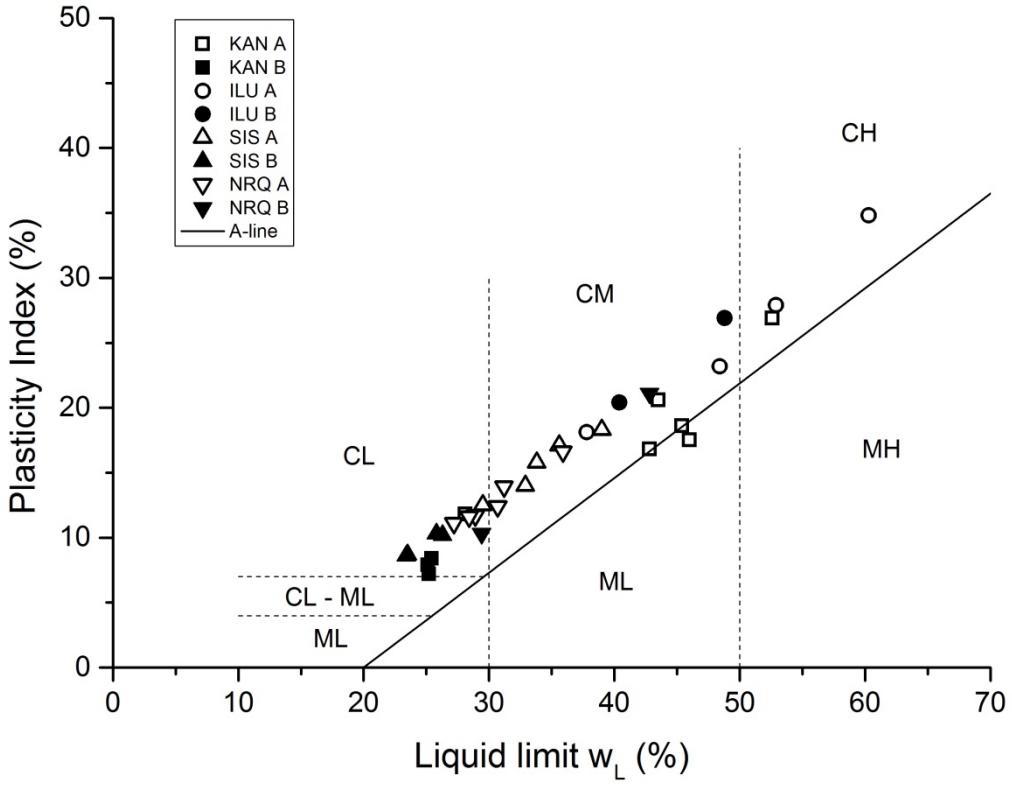

Figure 5. Casagrande plasticity chart modified according to Department of the Navy (1986) and Larsen et al. (1995). CL = clay of low plasticity, CM = clay of medium plasticity, CH = clay of high plasticity, ML = silt of low plasticity and MH = silt of high plasticity

Figure 5 shows the plastic properties of the Greenlandic sediments plotted in a modified version of Casagrande's plasticity chart (Casagrande, 1948; Department of the Navy, 1986; Larsen et al. 1995). Most samples plot well above the A-line, with 13 samples classified as clay of low plasticity (CL), 13 samples as clay of medium plasticity (CM), and 3 samples classified as clay of high plasticity (CH) according to the unified soil classification system (Department of the Navy, 1986). 3 samples from KAN A plot around the A-line and would be classified as borderline between clay of medium plasticity (CM) and silt of low plasticity (ML).

As the investigated sediments are all of marine origin, it is assumed that the pore water would initially have had a composition equal to that of seawater. This is consistent with assumptions made by Roaldset (1972) for Norwegian glaciogene marine clays. The chemical composition of the

pore water has therefore been compared to the composition of average sea water as specified by Neumann and Pierson (1966). Figure 6 shows the pore water to sea water concentration ratios (in percent) for Na^+, K^+, Mg^{2+}, Ca^{2+} and Cl^-. In these diagrams, a value of 100 % (marked by a black line) would indicate a composition equivalent to that of sea water, while a value below 100 % indicates a depletion of the respective ionic species.

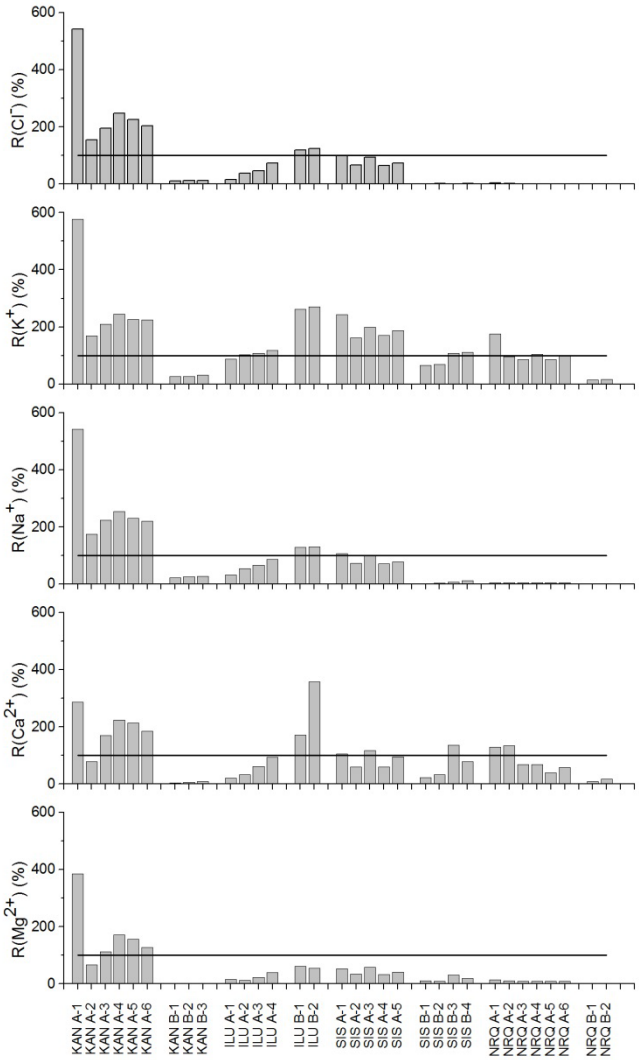

Figure 6. The pore water to sea water concentration ratios for Cl^-, K^+, Na^+, Ca^{2+} and Mg^{2+}. 100 % (black line) represents average sea water concentration as specified by Neumann and Pierson (1966).

4. Discussion

Overall properties of the sediments

Although the samples investigated here are distributed over a large geographical area, their overall grain size distributions and qualitative mineralogies were similar. They were characterised by a fine grain size distribution of 3-19 wt % in the < 0.2 µm (colloid) fraction, 18-69 wt % in the < 2 µm (clay) fraction and 71-100 wt % in the < 60 µm (silt) fraction. All samples contained quartz, feldspar, amphibole, biotite/illite, chlorite and expandable clay minerals and the primary minerals (e.g. quartz, feldspar and amphibole) were also observed in the colloid fraction. The higher total WPAR content of expandable minerals in the < 0.2 µm fraction compared to the 0.2-2 µm fraction of all investigated samples (see Table 3) suggest a mild weathering, where the < 0.2 µm fraction have been more weathered than the coarser and less reactive 0.2-2 µm fraction. The reduction of the WPAR content of illite in the < 0.2 µm fraction compared to the 0.2-2 µm fraction indicates that this mineral was altered into an expandable clay mineral, which would be consistent with conclusions made by Pederstad and Jørgensen (1985), Jensen (1965) and Jensen and Würtz (1974).

According to the Casagrande chart it is clear that the term “rock flour”, which has previously been used to describe the Greenlandic marine sediments (Jensen, 1965; Jensen et al., 1974; Heling, 1976), is generally not applicable to the samples investigated in this study, as most plot well above the A-line. Although they might have an origin as “rock flour” their current geotechnical properties combined with their mineralogy clearly define them as clays according to Guggenheim and Martin (1995). Furthermore, as their grain size distributions, mineralogies and plastic properties correspond well with those found for Canadian and northern Scandinavian marine clays (Gillott, 1971; Roaldset, 1972; Bentley and Smalley, 1978; Gillott, 1979; Locat et al., 1984; Pederstad and Jørgensen, 1985; Ramesh and d’Anglejan, 1995), the proper classification as clays should help align the nomenclature with these areas.

Origin of expandable clay minerals

The presence of the expandable clay minerals raises the question of their origin, for which there are several possibilities. They could have resulted from one or more of the following processes: pre-glacial or sub-glacial weathering of bedrock, weathering in the pro-glacial fluvial system, weathering under marine conditions or, for some, weathering after exposure to sub-aerial conditions.

Although Eberl (1984) argued that clay minerals in the polar regions are dominantly inherited rather than formed or transformed (i.e. produced from solution or by weathering of existing minerals), this seem unlikely for the samples investigated here. The geology of the investigated areas is dominated by igneous and high-grade metamorphic rocks and pre-glacial clay minerals would therefore most likely have resulted from surface or near surface weathering before the appearance of the Ice sheet. Cowton et al. (2012) estimated a current erosion rate of 4.6 ± 2.6 mm from beneath the Greenland Ice Sheet near Kangerlussuaq and concluded that Ice sheet margins were highly efficient agents of erosion during deglacial periods when melt water was abundant. It is therefore likely that any surface weathering would have been eroded at an early stage and is unlikely to explain the widespread occurrence and vast quantities of the fine-grained marine deposits and their similar composition with respect to expandable minerals regardless of geographical region.

Graly et al. (2014) conducted a study on chemical weathering under the Greenland Ice sheet based on bulk chemistry of the dissolved load in outlet water samples and concluded that silicate weathering mechanisms played an important role. Furthermore, several authors have found that glacial waters carry a higher proportion of K^+ compared to non-glacial waters (Anderson et al., 1997; Wimpenny et al., 2010). Newman and Brown (1969) found that mica edges that had been subjected to grinding or abrasion showed a higher rate of K^+ exchange than ordered edges (e.g.

cleavage planes). This observation is consistent with leaching of K^+ from e.g. illites in the glacial environment, which have been subjected to severe abrasion and grinding. Furthermore, this seems to suggest that chemical weathering is an active process underneath the ice, at least when water is present, i.e. under a warm-based glacier.

As the fine-grained sediments investigated in this study typically have low retention time in glacial rivers, it is unlikely that processes in this environment will have any significant influence on the weathering.

Bischoff and Ku (1970, 1971) and Siever et al. (1965) reported K^+ enrichment coupled with Mg^{2+} reduction in the pore water of deep sea sediment cores from geographically widespread localities in the Atlantic Ocean. Siever et al. (1965) suggest that the change in the K^+-Mg^{2+} ratio in pore water could be caused by dissolution of illite or K-feldspar combined with Mg^{2+} uptake by exchange in existing clay minerals. Bischoff and Ku (1971), however, found that although K-feldspar dissolution might occur in some sediments, it was not a general mechanism for K^+-enrichment. Powers (1957) investigated clay minerals from the James River estuary, Chesapeake Bay, and found that clay minerals in a marine depositional environment were more likely to adsorp Mg^{2+} than K^+ on to their surface. The K^+-Mg^{2+} ratio relationship therefore appears to be a global feature of recent marine sediments and implies that changes to the initially marine chemical composition of the pore water occur shortly after sedimentation.

Although chemical weathering under sub-aerial conditions in cold climates has been debated, several authors have reported evidence in favour of this (Pederstad and Jørgensen, 1985; Ramesh and d'Anglejan; 1995; Thorn et al., 2001). Pederstad and Jørgensen (1985) investigated a marine clay from Norway, which was deposited after the last glaciation and lifted above sea level approximately 8.5 ka BP. They found that trioctahedral illite in the upper part of the profile was transformed by subaerial weathering into vermiculite and mixed layer clays.

Chemical profiles

Based on the above discussion we rule out pre-glacial weathering and weathering in the pro-glacial fluvial system as like contributers, and find that the most likely scenarios are subglacial weathering, marine alteration or subaerial weathering. The samples investigated in this study allow us to focus on the last two scenarios. In order to clarify the observable processes in each of the two scenarios, the Cl^- concentration ratios and current geological settings of the investigated boreholes were divided into three distinct profile types: marine, depleted and enriched profiles:

Marine profiles

The marine profiles are observed in the two boreholes located below sea level, ILU B and SIS A, which both have Cl^- concentration ratios around 100 %. Although they are both classified as marine profiles, they show different trends in their chemistry. ILU B have Ca^{2+}, Na^+, and Cl^- concentration ratios above 100 %, whereas SIS A have ratios around or below 100%. The increased concentrations observed in ILU B likely originate from consolidation of the sediment. Geotechnical oedometer tests (Foged, 1979) corresponds to effective stresses from more than 30 m of overlying sediments, which have eventually been removed by erosion. The consolidation process could make clay act as a semipermeable membrane, draining water and concentrating ions in the sediment. As mentioned in section 2.1 the geological evidence for SIS A does not exclude the possibility that these samples have been exposed to short periods of subaerial conditions and leaching, which could account for the slightly lower concentration ratios. Both the ILU B and SIS A profiles had ratios above 100 % for K^+ and the ratios appear to be elevated compared to other ions[1], indicating that K^+ ions have been supplied to the pore water from another source than sea water after deposition. Oppositely, both profiles had ratios below 100% for Mg^{2+}, and the ratios

[1] Ca^{2+} concentrations in the samples measured by Foged (1979) might be overestimated, see Appendix A

appear to be lower compared to other ions, indicating that Mg^{2+} have been depleted from the pore water.

Enriched profile

An enriched profile is observed in the borehole KAN A, where the Cl^-, K^+, Na^+, Ca^{2+} and Mg^{2+} concentration ratios are higher than 100 % for most samples. The profile is a result of the arid climate, high net evaporation and continuous capillary uptake of water of marine salinity as described in section 2.1. Although KAN A is situated in the continuous permafrost zone, and temperatures as low as -3.6°C were observed in the deeper samples immediately after extraction, the samples were not technically frozen (i.e. they contained liquid pore water). This is consistent with the observed high salinities causing a freezing point depression of 4.2°C for the pore water composition of the lower samples, calculated using FREZCHEM (Marion, 2010).

Depleted profiles

The depleted profiles are observed in boreholes KAN B, ILU A, SIS B, NRQ A and NRQ B, where all samples have Cl^-, Na^+ and Mg^{2+} concentration ratios lower than 100 %, due to leaching by percolating fresh water. Also Ca^{2+} concentration ratios are generally lower than 100 %, except a few cases, which are most likely explained by overestimation of Ca^{2+} in the porewater[1]. The samples from ILU A show an increasingly marine signature with depth, with ion ratios approaching 100 %, indicating that leaching was halted at an early stage, most likely due to the formation of ice-rich permafrost after the Holocene optimum (5-8 ka BP). The KAN B and SIS B profiles are affected by ice-rich permafrost as well, but do not show trends similar to ILU A. The KAN B and SIS B samples are, however, more coarse grained than the ILU A samples and thus more permeable to water and susceptible to leaching, before ice-rich permafrost was formed. The K^+ concentration ratios are close to or above 100 % for the ILU A, SIS B and NRQ A samples and for the KAN B and

NRQ B samples they appear to be elevated in comparison to other ion ratios. This suggests that K^+ have been supplied to the pore water after deposition.

Weathering in the marine and subaerial environments

The elevated K^+ ratios compared to the other ion ratios are a general trend for all three profile types. In most of the profiles the elevated K^+ ratios are coupled with reduced Mg^{2+} ratios. In summary, the findings of previous authors (Bischoff and Ku (1970, 1971); Powers (1957); Siever et al. (1965)) support our findings that weathering processes occur after deposition of the marine sediments, and that K^+ is released to the pore water while Mg^{2+} is consumed.

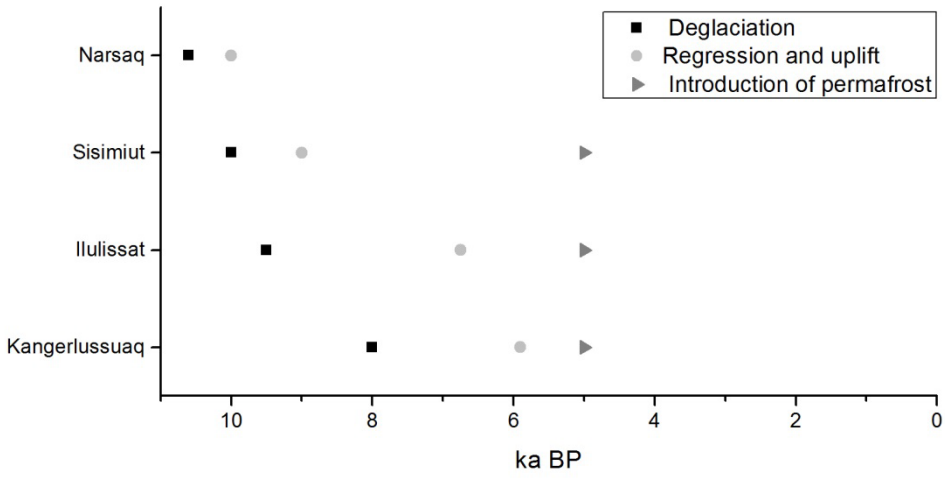

Figure 7. Approximate timing of the deglaciation, regression, uplift and introduction of permafrost in the four towns (see section 2.1 for references)

Figure 7 demonstrates the approximate timing of the events, which could have influenced the weathering properties of the depleted profile samples. NRQ A and NRQ B have been exposed to the longest periods of leaching after uplift and have not been subjected to permafrost during this period. The two boreholes generally have a higher WPAR content of expandable minerals in both the 0.2-2 µm and < 0.2 µm fractions than observed at other localities, which indicate that they are

more weathered. This effect is also observed, although less pronounced, for the SIS B samples, which have been exposed to a shorter period of leaching. The chemical evidence also supports the hypothesis that the samples from these boreholes are more weathered than the other investigated boreholes, as in general, the most depleted samples (in respect to marine pore water and Cl^-) are also the most enriched in K^+. The low K^+ concentration ratio observed for the NRQ B samples is likely explained by the coarser grain size distribution which would signify easier transport of fluids through the sediments, and thereby more effective leaching of all elements.

The ILU A samples, which have been exposed to a shorter period of leaching, than the NRQ and SIS B samples, show an increasingly marine signature with depth both in the < 0.2 µm WPAR mineralogy and pore water ion concentration ratios. The signature indicates that leaching of marine pore water from these samples has been incomplete, likely caused by the introduction of ice-rich permafrost. The signature is, however, less pronounced for K^+ compared to the other ions, again indicating that the most depleted samples (in respect to marine pore water and Cl^-) are also the most enriched in K^+, i.e. weathered.

Despite being exposed to the shortest period of leaching before the introduction of permafrost, KAN B do not demonstrate the same increasingly marine signature with depth as ILU A, which indicate that these samples have been completely leached of their marine pore water before the introduction of ice-rich permafrost. The difference between the two can possibly be explained by the coarser grain size distribution of the KAN B samples, which, similar to what was observed for the NRQ B samples, would make the leaching more effective.

When comparing the WPAR mineralogy of the most depleted samples (NRQ A, NRQ B and SIS B) to the marine (ILU B and SIS A) and enriched (KAN A) samples, it appears that the depleted samples generally have a higher content of expandable clay minerals, which suggests that the depletion of marine pore water enhances weathering.

The introduction of ice-rich permafrost in the KAN B, SIS B and ILU A samples appear to slow down or stop the weathering processes. This is especially clear in the case of the ILU A samples, where the marine signature with depth is preserved even after being subaerially exposed for > 6.5 ka.

In summary, we argue that expandable clay minerals are formed as weathering products in subglacial, marine and/or subaerially exposed environments. Our data allowed us to study the last two scenarios and indicate that the subaerially exposed samples, which have been leached by percolating fresh water, are more weathered than the marine samples. Furthermore, ice-rich permafrost appears to slow down the weathering process.

5. Conclusion

The combined dataset of the present study has shown the fine-grained marine sediments from western Greenland contain clay minerals and have plastic properties, which generally classify them as clays according to AIPEA and CMS nomenclature (Guggenheim and Martin, 1995).

Although the samples investigated here are distributed over a large geographical area, their grain size distributions and qualitative mineralogies are very similar. Furthermore, these properties are also very similar to those found for Canadian and northern Scandinavian marine clays.

The expandable clay mineral content is direct evidence of chemical weathering and was most likely formed during glacial weathering of bedrock, after deposition in sea water or by sub-aerial weathering. Furthermore, there appears to be an enrichment of K^+ in the pore water of the sediments, which could be caused by alterations of e.g. illite.

There appears to be a correlation between the exposure to leaching and WPAR distribution of expandable clay minerals, i.e. the samples with the highest WPAR distributions of expandable clays are also the samples which have been subjected to the longest periods of leaching. For sub-aerial sediments, the formation of ice-rich permafrost may halt the leaching of the marine pore

water, however, most of the investigated profiles contained low concentrations of ions compared to sea water irrespective of their exposure to permafrost.

Acknowledgements

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Appendix A

A review of the methodology applied by Foged (1979, 1978, 1975) to the samples from Ilulissat (ILU-A and ILU-B), Sisimiut (SIS-A and SIS-B) and Narsaq (NRQ-A and NRQ-B).

Particle size distributions

Approximately 40 g of each sample was dispersed in 0,3 L of distilled water. For samples with a high residual salinity, the water was replaced until dispersion (based on visual inspection) was achieved. 12 mL of 0,1 M $Na_4P_2O_7$ peptisator was added in order to ensure complete dispersion. The solution was split into two fractions by wet sieving through a 63 µm sieve. The > 63 µm fraction was dried and the grain size distribution was determined by sieving according to BS 1377:1975. The grain size distribution of the 2 µm < Ø < 63 µm and Ø < 2 µm fractions were determined by sedimentation using the pipette method according to Andreasen (1939) and by use of the spherical diver method by Berg (1940), respectively.

Atterberg limits and loss on ignition (LOI)

For all samples, the natural water content was measured by drying the sample at 105-110°C for more than 14 hours, and determined as the percentage mass loss compared to dry mass. This procedure follows the principles of CEN ISO/TS 17892-1:2004.

The liquid limits of the samples were determined by use of the Casagrande method in accordance with BS 1377:1975 and ASTM D423-66:1972. Plastic limits were determined by the rolling method in accordance with BS 1377:1975 and ASTM D424-54:1971.

The loss on ignition (LOI) was determined as the weight loss between 110°C and 600°C and was measured after heating at 600°C for 4 hours.

Mineralogy

Sample preparation and measurement

The samples were dispersed and the < 2 µm fraction was separated by use of the elutriation technique (Jensen and Hansen, 1961). The < 0.2 µm was separated by centrifuge and precipitated by use of NaCl. Both fractions (0.2- 2 µm and < 0.2 µm) were subsequently saturated with $MgCl_2$ and orientated samples were prepared by use of the smear method.

Pre-treatments of the sediments in order to remove organic matter, $CaCO_3$ and Al and Fe-Al oxides, before conducting mineralogical investigations, were not applied. Due to low organic C (see Table 3) and $CaCO_3$ contents in the investigated samples, it was considered unnecessary to apply treatments to remove these, especially as the treatment of removing $CaCO_3$ by HCl had been reported to cause break down of chlorites (Brindley, 1972). Fe-oxides were identified in several of the Greenlandic samples, however, Foged (1979) and Roaldset (1972) concluded that a much applied method for the removal of Fe-oxides, the dithionite-citrate treatment described by Mehra and Jackson (1960), did in fact alter the clay mineral compositon of the glaciogene marine clays from Greenland and Norway. Foged concluded that the diothionite-citrate treatment attacked the stability of the trioctahedral illite and found an increase in the vermiculite content coupled with a decrease in the illite content in the treated sample compared to the untreated sample. These alterations were not explained by any other characteristics of the clay and illites, and the treatment for Fe-oxides were therefore also discarded.

The samples were analysed on a Philips PW1050 X-ray diffractometer, equipped with a graphite monochromator and an energy dispersive detector. Cu K_α radiation was generated using 40 kV and 24 mA. A scan speed of 0.008 °2Θ/s were used for data collection.

The (060)-reflection of illite was measured on unorientated powder samples prepared for selected samples. The measurements were obtained by Guinier camera, using graphite monochromated Cu $K_{\alpha 1}$ radiation generated at 40 kV and 24 mA.

Mineral identification and semi-quantitative distribution:

The mineral phases were identified according to their behaviour after treatments of air-drying, ethylene glycolation (vapour) at room temperature for at least 48 hours and heating at 350°C and 550°C, respectively, for a minimum of 2 hours (Brown and Brindley, 1980). The phases studied were montmorillonite (in this study termed smectite), vermiculite, illite (as defined by Grim et al. (1937)), mixed layer clay minerals (regular) and unidentified expandable clay minerals (random mixed layer), chlorite and kaolinite. The weighted peak area ratio (WPAR) method was used for semi-quantitative analyses of the mineral phase distribution in the samples. The intensity weighing scheme and formulas for the WPAR calculations are identical to the ones given in Table 2. The areas of the individual peaks were determined by integration.

Chemistry

5 g of undried material from each sample was dispersed four times by ultrasonics in 20 mL distilled H_2O and was then collected in a 100 ml flask and filtrated through a 0,6 µm filter by use of vacuum pumping. Cl^- in the solution was analysed by titration with $Hg(NO_3)_2 \cdot H_2O$ using diphenylcarbazone as an indicator according to the method described by Schales and Schales (1941). Mg^{2+} and Ca^{2+} were analysed by atomic absorption spectroscopy (AAS) using an additive of $LaCl_3$ in order to avoid the formation of coordination complexes. It should be noted that Ca^{2+} concentration might be overestimated, as the pH was not buffered during sample preparation and dissolution of $CaCO_3$ might therefore have contributed to give a higher Ca^{2+} concentration. Na^+ and K^+ were analysed by

flame photometry. Results were corrected for initial content of pore water, i.e. natural water content.

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Appendix 2

Description: Unpublished paper

Title: Screening of marine sediments from three Greenlandic localities for use in the production of bricks

Screening of marine sediments from three Greenlandic localities for use in the production of bricks.

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Abstract

Marine sediments from near the Greenlandic towns of Ilulissat, Kangerlussuaq and Sisimiut were assessed as raw materials for brick production with the purpose of identifying the most suitable occurrence. The screening method used in this work includes both analysis of the sediment characteristics (e.g. grain size distributions, Atterberg limits and mineralogy) and properties of fired test brick pellets (e.g. open porosity, water absorption and density). The samples and brick pellets were, in general, very similar in respect to the investigated properties. The samples from Kangerlussuaq and Sisimiut were, however, precluded due to a high salt content and low plastic properties, respectively, and the sample from Ilulissat was therefore found to be the most suitable.

Keywords: marine sediments, screening, bricks, Greenland

1. Introduction

Vast occurrences of fine-grained marine glaciogene sediments, which have been uplifted above sea level, are found throughout the former glaciated regions of the northern hemisphere. In West Greenland these occurrences have not been exploited, but are often found in close vicinity to towns, which make them easily accessible for industrial purposes. Recent research from other

areas of the world has established the potential of producing bricks from marine sediments (Baruzzo et al., 2006; Hamer and Karius, 2002; Mezencevova et al., 2012; Romero et al., 2008; Salim et al., 2012) and in Greenland a pilot-study of one occurrence near the town of Kangerlussuaq (Villumsen, 2013) proved that bricks could also be produced from sediments originating from here.

Belmonte et al. (2014) established that although the Greenlandic marine sediments appear to be very homogeneous in grain size distribution and mineralogy, there were still detectable differences, e.g. in the plastic properties and the content of salts. These differences makes it difficult to choose the most suitable occurrence for brick production and a test production is therefore needed in order to confirm the resource potential and to optimize production conditions. An initial screening process can often help to identify the most promising resources and thereby save money and time compared to the larger scale test productions. Several authors have demonstrated the usefulness of small scale brick pellets for initial testing of ceramic properties (Isik and Messer, 1983; Romero et al., 2008; Torres et al., 2009) and Romero et al. (2008) used brick pellets to characterise the sintering behaviour of marine sediments from three localities in Spain. In this paper, a similar screening method was applied to marine sediments from three different geographical localities in Greenland in order to identify the one with the highest potential. The three sediments were rated on the basis of both sediment characteristics and on the properties of the fired brick pellets. The sediment characterization included determination of grain size distribution, plastic and liquid limits and calculation of the plasticity index. The plasticity parameters are important for evaluating the possible industrial methods for forming the bricks prior to firing. Some key characteristics used generally to evaluate possible materials for brick production were also measured (the contents of carbon, sulphur and chloride and the overall mineralogy). The properties of the fired brick pellets were investigated through firing shrinkage,

apparent density, open porosity and water absorption. These properties are routinely measured in order to evaluate the firing process and assess the durability of fired bricks (ASTM C62-13a; Bloodworth et al., 2007; Brick Industry Association, 2006a; Karaman et al., 2006; Zhang, 2013).

2. Materials and Methods

2.1 Materials

Three samples of marine sediments were collected from areas within or near the West Greenlandic towns of Ilulissat, Sisimiut and Kangerlussuaq. The three towns are located at distances of approximately 130 - 280 km from each other and the collected samples therefore represent different geographical areas in West Greenland. The occurrences were chosen due to their accessibility from existing infrastructure, which could be a decisive factor for future brickworks. The sampling depth was 0-1 m for all samples. For the Ilulissat and Sisimiut samples, 0.2 m and 0.8 m, respectively, of organic-rich overburden had to be removed in order to reach the marine sediments.

2.2 Raw material analyses

The grain size distributions were analysed by a combination of wet sieving and sedimentation (using the hydrometer method) in accordance with DS/CEN ISO/TS 17892-4 (2004). The natural water content was determined according to DS/CEN ISO/TS 17892-1 (2004). The liquid limit was measured using the fall-cone method and the plastic limit was determined by the rolling method in accordance with DS/CEN ISO/TS 17892-12 (2004). For measurements of Cl^- concentration, 4 g of dried material from each sample was dispersed in 50 ml distilled H_2O and filtrated through a 0.45 µm filter by use of vacuum pumping. The Cl^- concentration was analysed by ion chromatography (IC) and the obtained results were corrected for initial content of pore water, i.e. natural water

content. Total carbon (TC) and sulphur (S) were measured using the combustion Infrared detection method on a LECO CS-200. The bulk mineralogy was analysed by X-ray powder diffraction (XRPD) on a X'Pert PRO diffractometer, using Cu Kα radiation generated at 45 kV and 40 mA. The three measured diffraction patterns were matched with the same reference minerals from the ICDD PDF-2 (2009-release) database and semi-quantitative analyses were obtained using the RIR (ratio-intensity-ratio) method integrated in the X'Pert software.

2.3 Pellet production

Materials for pellet production were dried at 105°C for at least 24 hours. The dried materials were lightly disaggregated using a mortar and pestle and larger visible rock fragments were removed. Afterwards, 2 g of material was placed in a small vial and distilled water was added in order to obtain a moisture content of 10%. The vials were sealed and the materials were left overnight to absorb the moisture. Pellets were produced by uniaxial compression in a purpose-built pellet mould placed in an automated Instron 6025 press. A maximum load of 14.76 kN (equivalent to a pressure of ~ 47 MPa) was chosen in accordance with Torres et al. (2009) and a compression rate of 0.25 mm/min were applied. The initial diameters and heights of the pellets were measured by a precision micrometer scale gauge. The pellets were dried at 105°C for 24 hours before the diameters and heights were measured again. The pellets were fired at 1000°C for 1 hour in a Vecstar laboratory Furnace, using an average heating and cooling rate of 6.8°/min ± 2.5°/min and 1.9°/min ± 0.5°/min, respectively. The maximum temperature was chosen in accordance with Torres et al. (2009) and because temperatures in the range of 900-1100°C, are standardly applied for initial investigations on bricks (Bloodworth et al., 2007; Dondi et al., 1997a,b; Zhang, 2013). After cooling to approximately 200°C, the pellets were removed from the furnace and placed in a desiccator in order to cool to room temperature before diameters and heights were measured again.

2.4 Test of pellets

The porosity, density and vacuum water absorption were determined following the principles of procedure Ti-B-25 (1983) by the Danish Technological Institute. The procedure is briefly described in the following. Samples were dried at 105°C, cooled to room temperature in a desiccator and weighed (m_{dry}). The dried samples were then placed in a desiccator under vacuum for approximately 3 hours. After 3 hours, deionised water at room temperature was led into the desiccator, so that samples were completely submerged. Vacuum was maintained for this condition for 1 hour. Hereafter, air was let into the desiccator and the submerged samples were left at atmospheric pressure overnight. The water saturated samples were weighed in water (m_{sw}) and in air after wiping excess water of the sample surface (m_{sa}). The different parameters were calculated as following:

The open porosity $= \frac{m_{sa}-m_{dry}}{m_{sa}-m_{sw}} \cdot 100\%$

Apparent (bulk) density $= \frac{m_{dry} \cdot \rho_w}{m_{sa}-m_{sw}} \cdot 100\%$, where ρ_w is the density of water at room temperature.

Vacuum water absorption $= \frac{m_{sa}-m_{dry}}{m_{dry}} \cdot 100\%$

3. Results

3.1 Sediment characteristics

The grain size distributions are presented in Figure 1 and although the Sisimiut sample is slightly coarser than the others, they are generally similar. All samples are dominated by grain sizes < 60 µm (silt and clay fractions) and the < 2 µm fraction (clay fraction) constitutes between 34-44 wt %.

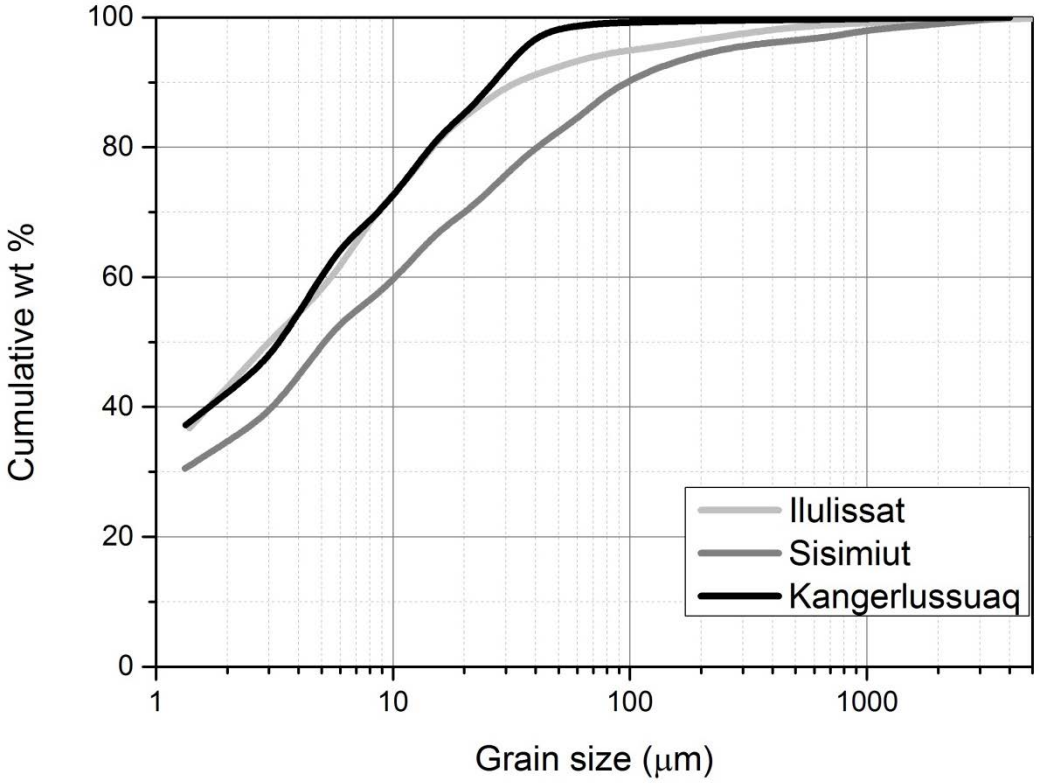

Figure 1. Grain size distributions of the investigated sediments.

The results of the geotechnical investigations are shown in Table 1 and the TC, S and Cl^- analyses are presented in Table 2. According to the Unified Soil Classification System (ASTM D2487-11, 2011), the samples from Ilulissat and Kangerlussuaq would classify as lean clays and the sample from Sisimiut as inorganic silt, based on their liquid limits, plasticity indices and low organic carbon content (which is inferred from the low TC).

Sample	Natural Water Content (%)	Plastic Limit (%)	Liquid Limit (%)	Plasticity Index
Ilulissat	23.5 ± 1.7	20.5 ± 0.9 (19-26)	30.1 (38-60)	9.6 (18-35)
Sisimiut	17.2 ± 0.6	15.1 ± 0.1 (15-16)	18.7 (23-26)	3.6 (9-10)
Kangerlussuaq	31.0 ± 0.6	19.5 ± 0.7 (16-29)	32.3 (28-53)	12.8 (12-27)

Table 1. Geotechnical properties of the investigated sediments. The values in parentheses are from Belmonte et al. (2014) and Foged (1979) and were measured on clay samples in near vicinity of the ones studied here.

Sample	TC (wt %)	Sulfur (wt %)	R(Cl^-) (%)
Ilulissat	1.42 ± 0.16	0.09 ± 0.03	0.90 ± 0.01
Sisimiut	0.17 ± 0.00	0.06 ± 0.08	0.86 ± 0.06
Kangerlussuaq	0.02 ± 0.00	0.03 ± 0.00	60.52 ± 1.07

Table 2. The total carbon content (TC), sulphur content (S) and chloride concentration ratio (R(Cl^-)) of the three investigated samples.

As Belmonte et al. (2014) and Foged (1979) investigated the plastic properties of marine sediments from Kangerlussuaq (locality KAN A, located approximately 2 km from the sampling locality in this study), Sisimiut (locality SIS B, located approximately 2 km from the sampling locality in this study) and Ilulissat (locality ILU A, located approximately 0.6 km from the sampling locality in this study), these values are included in Table 1 for comparison. As all the samples are of marine origin, the results of the Cl^- analyses are presented as percentage ratios of the chloride concentration in the pore water (ppm) divided by the chloride concentration in average sea water (ppm). A chloride concentration ratio (R(Cl^-)) of 100 % represents average sea water concentration as specified by Neumann and Pierson (1966). The chloride concentration ratios of the Ilulissat and Sisimiut samples are 0.90 % and 0.86 %, respectively, which indicate that although both samples had a marine origin, their pore waters have since been almost completely depleted of their original chloride content, due to leaching. The R(Cl^-) value of 60.52 % for the Kangerlussuaq samples, demonstrates that this sample has been subjected to some leaching but still contains a high concentration of chloride from the marine environment. The high salinity of the Kangerlussuaq occurrence was also observed by Belmonte et al. (2014) who found that the continuous capillary uptake of marine fjord water and the arid climate in the Kangerlussuaq area, which resulted in high net evaporation, was the cause of this.

The results of the mineralogical investigations are shown in Table 3. The RIR method applied in this study give a semi-quantitative evaluation of the mineral content in the samples, however, the results show that the overall distributions of the investigated major phases are very similar in spite of the geographical separation between the samples.

Sample	Quartz (%)	K-feldspar (%)	Na,Ca – feldspar (%)	Biotite (%)	Amphibole (%)	Chlorite (%)
Ilulissat	25	16	36	10	7	7
Sisimiut	19	12	42	11	11	5
Kangerlussuaq	20	16	38	9	10	7

Table 3. Mineralogical distribution (as determined by the RIR-method) of the major mineral phases in the three investigated samples.

3.2 Properties of brick pellets

The results of the measurements on the drying and firing shrinkages, open porosity, apparent density and vacuum water absorption are seen in Table 4.

Sample	Drying shrinkage (%)	Firing shrinkage (%)	Open porosity (%)	Apparent (bulk) density (kg/m^3)	Vacuum water absorption (%)
Ilulissat	0.4 ± 0.1	0.3 ± 0.1	28 ± 0.9	1970 ± 23.4	14 ± 0.6
Sisimiut	0.1 ± 0.1	-0.1 ± 0.1	22 ± 1.3	2130 ± 30.3	10 ± 0.7
Kangerlussuaq	0.0 ± 0.1	0.2 ± 0.2	31 ± 0.6	1870 ± 14.7	16 ± 0.5

Table 4. The drying shrinkage of the dried pellets and firing shrinkage, open porosity, apparent (bulk) density and vacuum water absorption of the fired pellets.

4. Discussion

Grain size and plasticity of the samples

The samples from this study all had plastic limits within the range found by Belmonte et al. (2014) for samples from the same geographical area. For the Ilulissat and Sisimiut samples the liquid limits and plasticity indices were lower compared to the values reported by Belmonte et al. (2014). However, these differences can most likely be explained by local variations, e.g. bands of coarser grained materials in the sediments. The Kangerlussuaq and Ilulissat samples were generally similar with respect to grain sizes and plastic properties. In comparison to this the Sisimiut sample had a coarser grain size distribution and lower plastic limit, liquid limit and plasticity index. The comparative coarser grain size of the Sisimiut sample in this study was also observed for the Sisimiut samples in the study by Belmonte et al. (2014), which may indicate a more general trend.

Macedo et al. (2008) reported typical values of plastic limits, liquid limits and plasticity indices in the range of 15-30%, 30-60% and 10-30, respectively, for clays used in red ceramic bricks. Compared to this, all the Greenlandic samples fall within the ranges of the plastic limits, the Kangerlussuaq and Ilulissat samples fall within the ranges of the liquid limit and the Kangerlussuaq sample falls within the limits of the plasticity indices. The slightly lower plasticity index of the Ilulissat sample (9.6) is however not critical, as Karaman et al. (2006) reported a plasticity index of 9 for a brick clay from Turkey. Based on the plastic properties of the Greenlandic sediments, the extrusion or dry pressing methods would be preferable compared to the soft-mud method (Brick Industry Association, 2006a). The very low liquid limit and plasticity index of the Sisimiut sample make this material suitable for dry pressing only.

Porosity, density, shrinkage and water absorption of the pellets

Bulk densities of clay bricks, which are used in the building industry, have been reported to be in the range of 1510-2380 kg/m^3 (Dondi et al., 2004; Dondi et al., 2000; Hall et al., 1992) and open porosities in the range of 18.8-39 vol % (Dondi et al., 2004). The densities (1870-2130 kg/m^3) and open porosities (22-31 vol %) found in this study, are therefore acceptable. Romero et al. (2008) reported open porosities in the range of ~28-45 % for pellets of marine clay from Spain, which were compacted at 40 MPa and fired at 1000°C. The higher compaction pressure (47 MPa) of the Greenlandic pellets could be an explanation for the lower open porosity observed in this study (Isik and Messer, 1983). However, the differences in open porosity could perhaps also be explained by the differences in clay characteristics. According to the Brick Industry Association (2006,a) the size shrinkage due to drying and firing usually fall within the ranges of 2-4 % and 2.5-4 %, respectively. In this study, both drying and firing shrinkages are below 0.5 %. For the Sisimiut pellets, a net negative firing shrinkage was observed, indicating a slight expansion of the pellets during firing. The slight expansion could be caused by the presence of small amounts of calcite or

sulphides which liberate CO_2 and SO_2 during firing and have been known to cause bloating (Riley, 1951). Although neither calcite nor sulphides were detected by the XRD, both minerals have previously been identified in minor quantities from Greenlandic marine clay from other localities (Foged, 1979) The overall shape and coherency of the pellets were, however, not affected by the slight expansion. Taylor (1991) stated that bloating can be caused by too-rapid firing, and extending the heating time could therefore possibly resolve this problem. Wilson et al. (1999) reported vacuum water absorptions of 6.5-22.5 % for engineering and facing bricks. The vacuum water absorptions measured in this study all fall within this range.

Mineralogy and chemistry

According to the British Geological Survey (Bloodworth et al., 2007), suitable clay for brick manufacturing should fulfil the following requirements: 1) contain an adequate proportion of non-plastic constituents, such as quartz, in order to prevent excessive shrinkage and deformation of the bricks during drying and firing 2) contain sufficient fluxing materials (e.g. Na and K compounds) which aid the vitrification at temperatures around 900-1100°C and 3) contain low levels of sulphur and carbon (< 0.1 % and 1.5 %, respectively) in order to control firing performance and emissions. The samples in this study contain between 19-25 % quartz and 52-54 % feldspar. Feldspars have good fluxing potentials, due to the presence of Na and K in the structure (Vieira and Monteiro, 2005). As the shrinkage and densities measured in this study are satisfactory, it must be concluded that the contents of quartz and fluxing material are also acceptable.

The S and TC values range from 0.03-0.09 wt % and 0.02-1.42 wt%, respectively, for the Greenlandic samples. The highest values are found for the Ilulissat samples, where especially TC is markedly different from the other samples. The higher TC could possibly be explained by shell fragments (calcite, although this was not observed by the XRD) or a higher content of organic carbon. However, all values were below the levels specified by Bloodworth et al. (2007).

The high salt content of the Kangerlussuaq sample could be problematic, as it might give rise to unwanted surface efflorescence or even mechanical weathering, i.e. spalling of the fired bricks (Brick Industry Association, 2006b). Furthermore, the presence of soluble chloride in the raw material can cause problems with kiln-related corrosion and emissions during firing, due to the formation of acid (HCl) (Voland et al., 2004). Additives such as $BaCO_3$ and lignosulphonate can reduce efflorescence by forming insoluble complexes; however, Andrés et al. (2009) found that chloride was still liberated as HCl during firing when these additives were used. The Brick Industry Association (2006b) recommends that soluble salts should be precluded from masonry materials when possible, in order to reduce or prevent efflorescence. For the pellets produced in this study, no salt efflorescence was observed. However, as the Kangerlussuaq sediment has a high salt content, it should be precluded, especially because there are optional samples with very low concentrations of salt, where the potential emission and efflorescence problems might be avoided. The evaluation of the investigated properties of the samples and pellets are summarised in Table 5. The Ilulissat clay and pellets have acceptable properties and the clay appear to be the best choice for further large scale investigations. The Sisimiut and Kangerlussuaq clay generally also have acceptable properties, however, their low plastic properties and high salt content, respectively, might be problematic in brick production.

Property	Ilulissat	Sisimiut	Kangerlussuaq
Plastic limit	+	+	+
Liquid limit	+	÷	+
Plasticity index	+	÷	+
TC	+	+	+
S	+	+	+
Cl^-	+	+	÷
Drying shrinkage	+	+	+
Firing shrinkage	+	+	+
Open porosity	+	+	+
Bulk Density	+	+	+
Water absorption	+	+	+

Table 5. Evaluation of the properties of the investigated samples and pellets. (+) indicates acceptable property and (÷) indicates problematic property.

5. Conclusion

The initial investigations on e.g. grain size distributions and plastic properties were similar to results reported in the literature for nearby localities and demonstrate that the samples of this study are generally representative of the investigated occurrences. However, a detailed mapping of the individual occurrences would be needed in order to establish their true variations and potential as brick clays. Although the samples and pellets, in general, have very similar properties, the Ilulissat sample was shown to have the highest potential as brick clay, and further studies should therefore focus on this sediment. The Kangerlussuaq and Sisimiut samples were precluded due to a high salt content and low plastic properties, respectively.

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Appendix 3

Description: Unpublished paper

Title: Use of a typical marine clay from Greenland for the production of red bricks

Use of a typical marine clay from Greenland for the production of red bricks

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Abstract

Uplifted occurrences of fine-grained glaciogene marine sediments are found throughout the northern hemisphere. In this study, a representative occurrence from Ilulissat, West Greenland was investigated as a potential resource for local brick production. The study comprised of three parts: 1) raw material characterisation based on grain size distribution, major element chemistry including total carbon, sulphur and chloride concentrations, mineralogy, morphology and Atterberg limits 2) production of test bricks and 3) testing of the bricks based on total shrinkage, water absorption, hygroscopic adsorption, open porosity, bulk density, compression strength, initial freeze-thaw resistance and mineralogy. The bricks produced proved to have excellent compression strengths, low open porosity and low water absorption. In conjunction with some of the other investigated properties, this indicates that this type of clay is highly suitable as a resource for bricks, which could be used in the arctic climate.

Keywords: Greenland, marine clay, bricks, material characterisation, material testing.

1. Introduction

Large occurrences of fine-grained glaciogene marine sediments were deposited after the last glaciation and some later uplifted above sea level due to isostatic movements. Today, they are found all over the formerly glaciated regions of the northern hemisphere, such as Canada,

northern Scandinavia and Greenland (Belmonte et al., 2014; Foged, 1979; Gillott, 1979; Locat et al., 1984; Ramesh and d'Angeljan, 1995; Roaldset, 1972; Rosenqvist, 1975). The sediments are dominated by the < 60 µm fraction (clay and silt fractions) and contain only minor amount of the 60-2000 µm fraction (sand fraction) (Belmonte et al., 2014; Foged, 1979; Gillott, 1979; Pederstad and Jørgensen, 1985). Their chemical compositions are dominated by SiO_2 (46-63 wt%), Al_2O_3 (14-23 wt%) and Fe_2O_3T (4-15 wt%) with an alkali-alkaline earth element content (i.e. the content of K_2O, Na_2O, MgO and CaO) of 12-16 wt % (Bentley and Smalley, 1978; Gillott, 1971; Locat et al., 1984; Pederstad and Jørgensen, 1985; Roaldset, 1972). They have a qualitatively similar mineralogy often consisting of feldspars, quartz, amphibole, mica/illite, chlorite, expandable clay minerals (e.g. smectite, vermiculite and mixed layer clays) and calcite and their fine and colloid fractions contain a high percentage of primary minerals such as quartz, feldspars and amphibole (Belmonte et al., 2014.; Foged, 1979; Bentley and Smalley, 1978; Gillott, 1971; Gillott, 1979; Locat, 1984; Pederstad and Jørgensen, 1985; Ramesh and d'Angeljan, 1995; Roaldset, 1972). The proportion of expandable clay minerals can vary considerably from region to region (Gillott, 1979). The organic carbon content is usually less than 1-2 wt % (Belmonte et al., 2014; Foged, 1979; Bentley and Smalley, 1978; Gillott, 1979 ; Locat et al., 1984) and the chloride content of the pore water (or pore water salinity) can vary according to the degree of leaching, i.e. exposure to percolating precipitation after uplifting, and to the current geological setting (Belmonte et al., 2014). In West Greenland these deposits are observed throughout the inhabited coastal regions and are, in most cases, easily accessible from towns and settlements.

Research world-wide has established that marine sediments are suitable as primary and secondary resources in the production of clay ceramics (Baruzzo et al., 2006; Hamer and Karius, 2002; Mezencevova et al., 2012; Romero et al., 2008; Salim et al., 2012) and although the Greenlandic deposits might not be directly comparable to those mentioned above, their abundance and fine

grained nature still suggest that they could be worth investigating as a suitable major resource for brick manufacturing.

Today, bricks are not commonly used nor produced in Greenland, where building materials such as wood and concrete are favoured (Bjarløv and Vladykova, 2011; Garcia; 2012; Madsen, 2000). However, as most of the construction materials used in the country, are imported, it is of great importance to investigate the potential for local production. In 2012 the value of imported goods for construction was approximately 62 million Euro, which makes up approximately 9 % of the total import to the country (Statistics Greenland, 2010 a). Furthermore, the current housing situation in Greenland is critical, due to long waiting lists and poor conditions of the buildings (Garcia, 2012; Naalakkersuisut, 2012). In 2012 the government of Greenland reported that about 70 % of the state-owned flats and terraced houses, which constitutes approximately 20 % of the housing in Greenland, were dilapidated (Naalakkersuisut, 2012; Statistics Greenland, 2010 b). In order to accommodate an increasing population and satisfy the demands for housing, it is therefore vital for the government of Greenland to construct new homes for the future.
Depending on the raw materials and production methods, bricks have several advantages that make them an interesting building material in arctic climate regions, such as Greenland: They are durable and require almost no maintenance in comparison to e.g. wood. Furthermore, they are fire-resistant and have good thermal and acoustic properties (Hornbostel, 1991).
In this paper, clay from the town of Ilulissat, West Greenland has been characterised and tested for use in brick manufacturing. Furthermore the technical suitability of the produced bricks as a local building material in the arctic climate has been evaluated by parameters such as compression strength, water absorption and initial freeze-thaw resistance.

2. Materials and methods

2.1 Materials and sampling

The investigated raw material was a fine-grained glaciogene marine sediment (MS) from the town of Ilulissat in central West Greenland. The area around Ilulissat was covered by ice during the last glaciation (Weichsel-Wisconsin) and marine sediments were deposited following the deglaciation of the area approximately 9600 years BP (Bennike and Björck, 2002). Around 6000-7000 years BP, the area was raised above sea level due to isostatic uplift (Rasch and Jensen, 1997). The bedrock in the area consists of reworked Archean granitic gneisses related to the Palaeoproterozoic Nagssugtoqidian orogen (Henriksen, 2005).

Approximately 150 kg of MS was sampled from a depth interval of 20 cm – 100 cm below surface. The material was roughly homogenised using a large shovel and distributed into 10 L plastic buckets, which were sealed and stored at room temperature until use. Furthermore, two additional raw materials (CR and CH) were used in the production of the bricks. CR is a fine grained residue from crushing of granitic rocks for aggregate production, which is produced at Betoncentralen, Nuuk, Greenland. CH is a ceramic product with high alumina and silicon content, known as chamotte. CH is currently not produced in Greenland but could be manufactured by crushing and milling of discarded bricks from a future production.

2.2 Methods

2.2.1 Characterisation of raw material

The particle size distribution of MS, CR and CH was determined by laser diffractometry. The samples were pre-dispersed in approximately 20 mL 0.005 M $Na_4P_2O_7$ using ultrasonics and then analysed by wet dispersion in deionized water on a Malvern Mastersizer 2000 laser diffractometer. The Mie theory was applied in the calculations of the particle size distribution using a real refractive index of 1.5 and an imaginary refractive index of 0.1.

Major-element analyses were determined using the X-ray fluorescence (XRF) technique on pressed powder tablets prepared from crushed bulk material. The tablets were analysed on a PanAlytical MagiX PRO, equipped with a Rh-tube and a gas flow detector. Excitation voltage and current were 60 kV and 53 mA. The loss on ignition (LOI) was based on weight loss between 105°C and 1000°C and was measured after heating at 1000°C for 2hours.

The mineralogy was analysed by X-ray diffraction (XRD) on a X'Pert PRO diffractometer, using Cu K_α radiation generated at 45 kV and 40 mA. In addition to bulk analyses, the < 2µm fraction of MS was separated by sedimentation and analysed as orientated samples after treatments of air-drying, ethylene glycolation (vapour) at room temperature for at least 48 hours and heating at 350°C and 550°C, respectively, for a minimum of 2 hours. The mineral phases in the < 2 µm fraction were identified according to Brown and Brindley (1980).

The morphology of the materials (mounted on carbon tape) was investigated by scanning electron microscopy (SEM) on a FEI Quanta 200.

Furthermore, total carbon (TC), sulphur (S), chloride (Cl^-) and Atterberg limits were measured for MS. TC and S were measured using the combustion infrared detection method on a LECO CS-200. The Cl^- concentration of the pore water was estimated by the following method: 4 g of dried MS was dispersed in 50 ml distilled H_2O for 24 hours and filtrated through a 0.45 µm filter by use of vacuum pumping. Cl^- (mg/L) was analysed by ion chromatography (IC) and the results were corrected for initial content of pore water, i.e. natural water content, in order to obtain the chloride concentration ratio ($R(Cl^-)$). The Atterberg limits (liquid and plastic limits) were determined in accordance with DS/CEN ISO/TS 17892-12 (2004). A 0.425 mm sieve was used in the determination of the liquid limit. An additional determination of the liquid limit was carried out using the Casagrande method (DIN 18.122-1, 1997).

2.2.2 Production of fired specimens

In order to evaluate the technical properties, a small-scale test production of fired specimens was carried out at the Danish brickworks, Petersen Tegl. The raw materials were untreated prior to production; however, larger particles (> 5 mm) were removed by hand if encountered during the mixing and moulding procedures. Three main compositions consisting of MS (in pure form or with 10% substitutions of either CR or CH) were prepared. Specifications for all test specimens are seen in Table 1.

Composition	MS (wt %)	CR (wt %)	CH (wt %)	Number of specimens	Coal furnace	Electrical furnace	Firing temperature (°C)
1A	100			2	X		1045-1055
1B	100			3		X	1070-1080
2A	90	10		3	X		1045-1055
2B	90	10		2		X	1030-1040
3A	90		10	3	X		1045-1055
3B	90		10	3		X	1030-1040

Table 1. Specifications for the test specimens.

The compositions were each mixed thoroughly in an industrial mixer and water was added in order to obtain a workable mass. The specimens were prepared by hand using the soft-mud method (Brick Industry Association, 2006) and a water lubricated wooden mould measuring 18.6*12*6 cm. A wooden piston was used to aid the removal of the mould from the formed specimens. After forming, the specimens were included in the normal production routine at Petersen Tegl. Here they were dried at maximum 70°C for 3-5 days. From each composition, half of the specimens were fired in a fully automated coal-fired tunnel kiln (reaching a maximum temperature of approximately 1045-1055°C) and the other half were fired in an electrical furnace (reaching a maximum temperature of either 1030-1040°C or 1070-1080°C). In both furnaces the heating and cooling rates were kept at approximately 20°C/hour, using a lowered rate of 4-

10°C/hour in the interval 520-620°C. The specimens were maintained (soaked) at maximum temperature for approximately 4 hours.

2.2.3 Characterisation of fired specimens

The total shrinkage of the specimens after firing was calculated as a percentage of the initial moulded length before drying. The water absorption due to capillary action was determined by the principles stated in DS/EN 772-11 (2011). The specimens were dried in a ventilated oven at 105°C for a minimum of 72 hours (3 days) in order to obtain dry mass. A plastic container of dimensions 22.5* 16.5*7.4 cm was equipped with two elongated metal supporting devices and filled with distilled water until the water level was approximately 5 mm above the supporting devices. The base of the specimens were placed on the supporting devices at time t0 and then taken out of the water and weighed regularly during the experiment. The experiments lasted up to 76 hours and the plastic containers were therefore closed with plastic lids in order to limit evaporation. For the following tests, two specimens from each composition were chosen and cut into five pieces designated with roman numerals I, II, III, IV and V (See Figure 1). Pieces I were used firstly for open porosity and bulk density measurements and secondly for strength tests. Pieces II and IV were used firstly for strength tests and secondly for mineralogy, pieces III were used for hygroscopic adsorption experiments and pieces V were used for initial freeze-thaw experiments. The open porosity and bulk density were determined according to the principles stated by DS/EN 772-4 (1998). The pieces (I) were dried at 105°C and pressures of <0.1 kPa and <1.5 kPa were used to eliminate the air contained in the open pores and to saturate the specimens with water, respectively. The compression strength was measured on a 60 tonnes hydraulic press on pieces (I, II, IV) dried at 105°C for a minimum of three days. For comparison, two regular red bricks, a WT131 (produced by Wienerberger) and a D46 (produced by Petersen Tegl), were included in the compression tests.

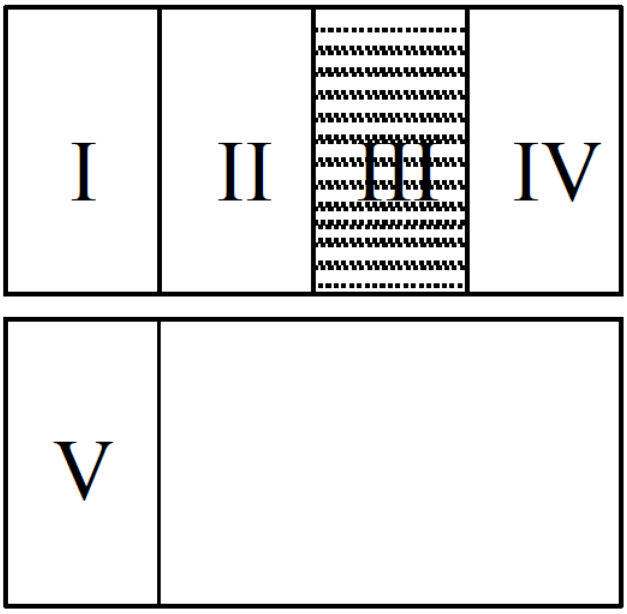


Figure 1. Two specimens from each composition were cut into five pieces for testing. Pieces III were further cut into square prisms.

The hygroscopic adsorption properties were determined according to the principles stated by ISO 12571 (2000) using the desiccator method. Small square prisms of approximately 1*1*2 cm and a weight of 3-11 g were cut from the pieces (III). The adsorption curves were established at a temperature of 20°C (± 2°C). Only one square prism from each sample was placed in the individual desiccators. Initial frost resistance were determined by freeze-thaw cycles over a period of 14 days. The pieces (V) were dried at 105°C and immersed in water according to DS/EN 12371 (2010). The apparent volume according to DS/EN 12371 (2010) and ultrasound velocity were determined before and after the freeze-thaw cycles on wet and dry pieces, respectively. The ultrasound velocity was determined by use of a PUNDIT apparatus. The wet pieces were wrapped in cling film and exposed to freeze-thaw cycles as listed in Table 2. Due to the small size of the pieces (average dimensions: width = 2.7 cm, length = 10.7 cm and height = 5.1 cm) and the short duration of the experiment, it was considered adequate to thaw the pieces in air, instead of in water as is

specified in the standard. The mineralogy of the fired specimens was investigated by XRD using the same equipment and operating conditions as described in 2.2.1.

	Temperature of freezing chamber (°C)	Time
Start of cycle	20	T_0
	-5	T_0 + 4 h
	-20	T_0 + 12 h
	-5	T_0 + 20 h
End of cycle	20	T_0 + 24 h

Table 2. The freeze-thaw cycles. The experiment included 14 consecutive cycles on wet pieces wrapped in cling film.

3. Results

3.1 Raw materials

The particle size distributions are presented in Figure 2. The d_{50} values were approximately 0.02 mm, 0.6 mm and 0.05 mm for MS, CR and CH respectively. MS was dominated by the < 60 µm fraction (clay and silt fractions), which was also characteristic of the clays from Canada, Norway and elsewhere in Greenland (Belmonte et al., 2014; Foged, 1979; Gillott, 1979; Pederstad and Jørgensen, 1985). Based on a d_{60}/d_{10} value of approximately 28, MS was classified as an unsorted sediment according to Larsen et al. (1995). The major element compositions and loss on ignition data are presented in Table 3. The major element composition of MS was equivalent to that of marine sediments from Canada and Norway (Bentley and Smalley, 1978; Gillott, 1971; Locat et al., 1984; Pederstad and Jørgensen, 1985; Roaldset, 1972). SiO_2, Al_2O_3 and Fe_2O_3T were the dominating oxides for all raw materials, however, CR had a high percentage of MgO (9.8 wt%), which made MgO more dominant than Fe_2O_3T for this raw material. The total amounts of alkali and alkaline earth elements (K_2O, Na_2O, MgO and CaO) were 12 wt%, 18.9 wt% and 4.3 wt% for MS, CR and CH respectively. Additional analyses on TC, S, Cl^- and Atterberg limits measured for MS are also shown in Table 3. The low TC was consistent with the low content of organic carbon

observed for other marine clays in Greenland, Canada and Norway (Belmonte et al., 2014; Foged, 1979; Bentley and Smalley, 1978; Gillott, 1979; Locat et al., 1984).

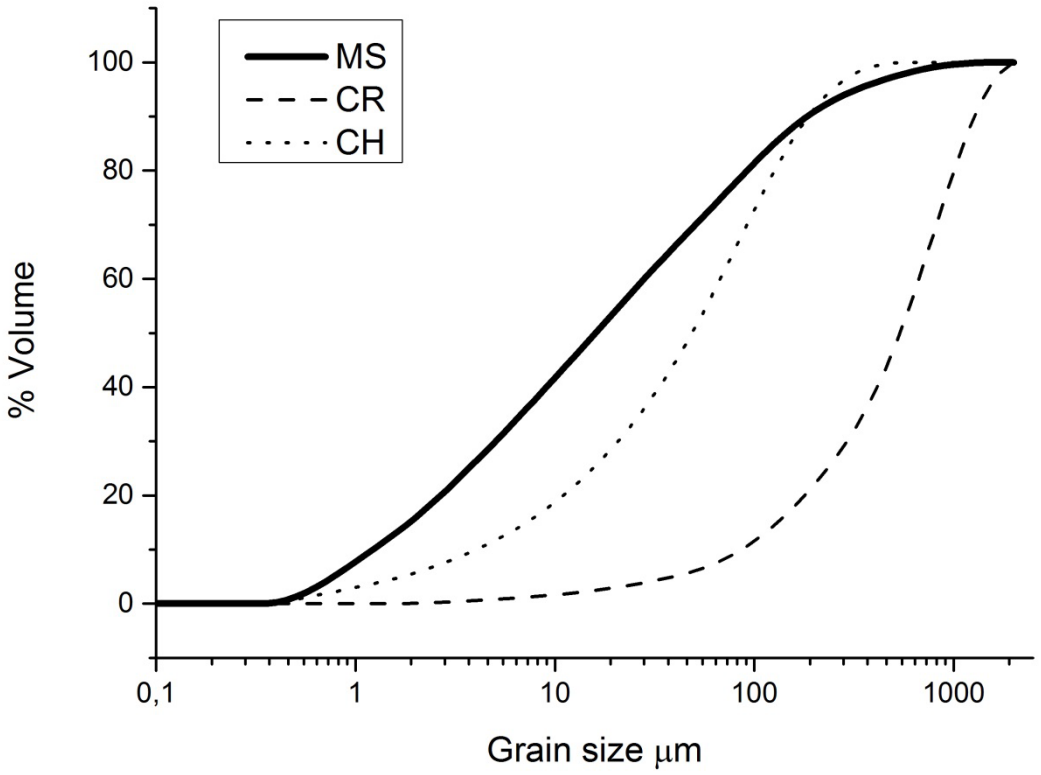


Figure 2. The particle size distributions of MS, CR and CH.

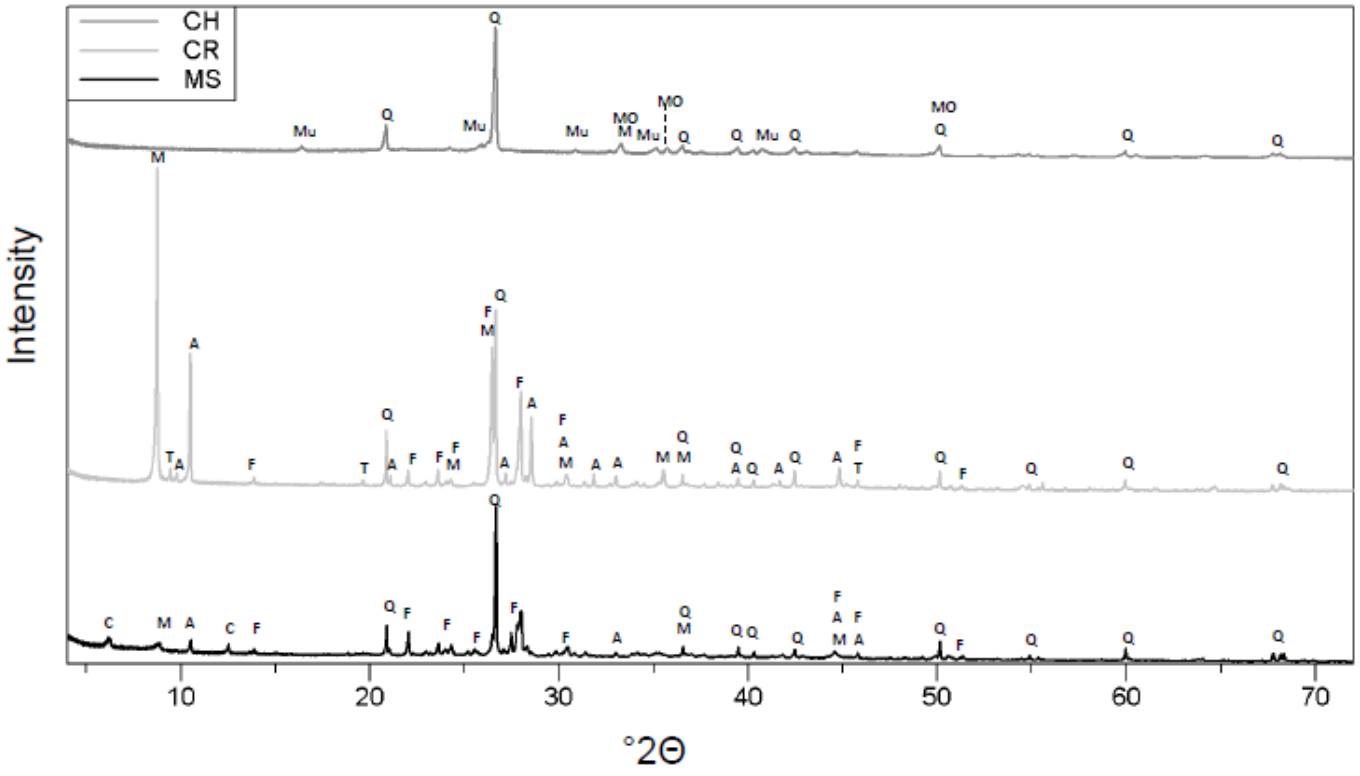
Furthermore the low TC also indicated a low content of $CaCO_3$, which combined with the relatively high amount of Fe_2O_3T contributed to the red colouration of the bricks after firing. As MS is of marine origin, the chloride concentration is also expressed as a percentage ratio of the chloride concentration measured in the pore water (ppm) divided by the chloride concentration in average sea water (ppm) according to Neumann and Pierson (1966). The chloride concentration ratio of MS is 0.9 %, which indicates that although MS had a marine origin the original salt content in the pore water has since been leached out. The results from the bulk mineralogy analyses of MS, CR and CH are presented in Figure 3A. The mica in MS was found to be dominantly trioctahedral (biotite-type), based on a (060) reflection around 1.54 Å and very weak peaks in the 1.50 Å region

(Brown and Brindley, 1980). For CR the octahedral coordination was less clear, but appeared to be more trioctahedral based on stronger reflections around 1.54Å than around 1.50Å. The high MgO wt % observed for CR (see Table 3) corresponds well with the abundant mica (biotite) found from the XRD investigations. For CR, the presence of pyroxene was also indicated from the diffraction data; however, due to peak overlapping this could not be clearly resolved.

		MS	CR	CH
Major elements and loss on ignition	SiO_2 (wt %)	61.12	61.77	66.94
	TiO_2 (wt %)	0.59	0.39	1.28
	Al_2O_3 (wt %)	14.52	10.63	19.22
	Fe_2O_3T (wt %)	6.48	6.45	7.80
	MnO (wt %)	0.08	0.11	0.06
	MgO (wt %)	3.52	9.67	0.99
	CaO (wt %)	2.36	3.64	0.33
	Na_2O (wt %)	3.31	3.11	0.43
	K_2O (wt %)	2.81	2.51	2.59
	P_2O_5 (wt %)	0.12	0.04	0.06
	LOI (wt %)	5.09	1.68	0.30
Carbon, sulphur and chloride	TC (wt %)	1.41 ± 0.18	N.M.	N.M.
	S (wt %)	0.10 ± 0.08	N.M.	N.M.
	Cl^- (mg/L)	3.29 ± 0.04	N.M.	N.M.
	$R(Cl^-)$ (%)	0.90 ± 0.01	N.M.	N.M.
Atterberg limits	Natural Water content (%)	23.5 ± 1.7	N.M.	N.M.
	Liquid Limit (%)	30.2 ± 0.2	N.M.	N.M.
	Plastic Limit (%)	20.5	N.M.	N.M.
	Plasticity Index	9.7	N.M.	N.M.
	Activity	0.23	N.M.	N.M.

Table 3. The major element distribution of MS, CR and CH and the total carbon content (TC), sulphur content (S), chloride concentration (mg/L), chloride concentration ratio ($R(Cl^-)$) and Atterberg limits of MS. N.M. = not measured.

A



B

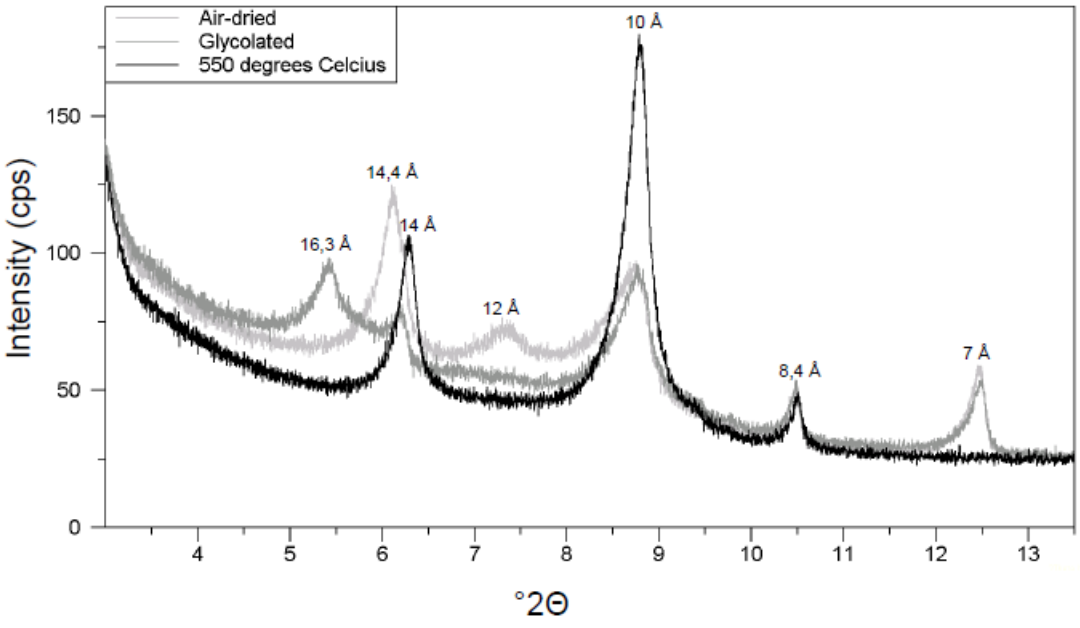


Figure 3. **A**: The bulk mineralogical composition of MS, CR and CH. A = amphibole, C = chlorite, F = feldspar, M = mica, MO = metal oxide (most likely an Fe-compound, e.g. hematite) Mu = Mullite, Q = quartz, T = talc. **B**: The clay mineral composition of MS.

For CH, cristobalite was observed from the diffraction data, but is not displayed in the figure due to the low intensity of the peaks. The result of the clay mineral analyses of MS is presented in Figure 3B. Expandable (smectitic) clay was identified based on the glycolated peak around 16.3 Å. Mixed-layer clay was identified based on the air-dried peak around 12 Å, which disappeared after glycolation and chlorite was identified based on the presence and increase of the 14 Å peak and the disappearance of the second order (7Å) reflection at 550°C. The shift from 14.4 Å to 14 Å from air-dried to 550°C could indicate the presence of vermiculite, although this would have to be confirmed by additional tests. Illite/biotite was identified from the 10 Å peak (before heating). The 8.4 Å peak seen in Figure 3B was assigned to amphibole, which together with quartz and feldspar peaks (not shown) confirmed the presence of primary minerals in the < 2 μm fraction. The overall mineralogy of MS (including both bulk and clay mineralogy) was consistent with that found for other marine clays in Greenland, Canada and Norway (Belmonte et al., 2014; Foged, 1979; Bentley and Smalley, 1978; Gillott, 1971; Gillott, 1979; Locat et al., 1984; Pederstad and Jørgensen, 1985; Ramesh and d'Angeljan, 1995; Roaldset, 1972). The morphology of the three raw materials as studied by the SEM revealed that CR and CH generally contained grains of euhedral textures, whereas the grains in MS had a more subhedral texture. Furthermore, the surfaces of larger grains in MS were generally covered by smaller grains with a flaky or powdery appearance, which were likely to be weathering products, e.g. clay minerals. This observation was less pronounced for CR and CH.

3.2 Fired specimens

The total linear shrinkages, bulk (apparent) densities and open porosities of the six different compositions are presented in Table 4. Composition 2B had the lowest linear shrinkage but also the highest open porosity and lowest bulk density, the opposite was true for composition 1B. The initial rate of water absorption (IRA) and capillary coefficients (k-values) are also presented in

Table 4. They generally correlate well, with the IRA values being about a factor ten higher than the k-values.

Composition	Total shrinkage %	Bulk (apparent) density kg/m^3	Open porosity %	IRA $kg/(m^2 \cdot min)$	k-value $kg/(m^2 \cdot s^{½})$
1A	13.8 ± 0.5	2350	4.9	0.170 ± 0.060	0.018 ± 0.005
1B	14.9 ± 0.4	2400	2.9	0.080 ± 0.050	0.008 ± 0.004
2A	11.0 ± 0.5	2260	16.8	0.300 ± 0.010	0.035 ± 0.003
2B	7.7 ± 0.8	1960	38.2	1.630 ± 0.060	0.183 ± 0.010
3A	13.8 ± 0.2	2360	9.3	0.080 ± 0.020	0.009 ± 0.003
3B	11.0 ± 0.4	2090	27.2	0.890 ± 0.180	0.094 ± 0.019

Table 4. The total shrinkage, bulk density, open porosity, initial rate of water absorption (IRA) and capillary coefficients (k-values) of the six tested compositions.

The compression strengths of the six compositions and of the WT131 and D46 bricks are plotted in Figure 4. The Greenlandic bricks showed remarkable strength up to 8 times higher than the values obtained for the regular bricks.

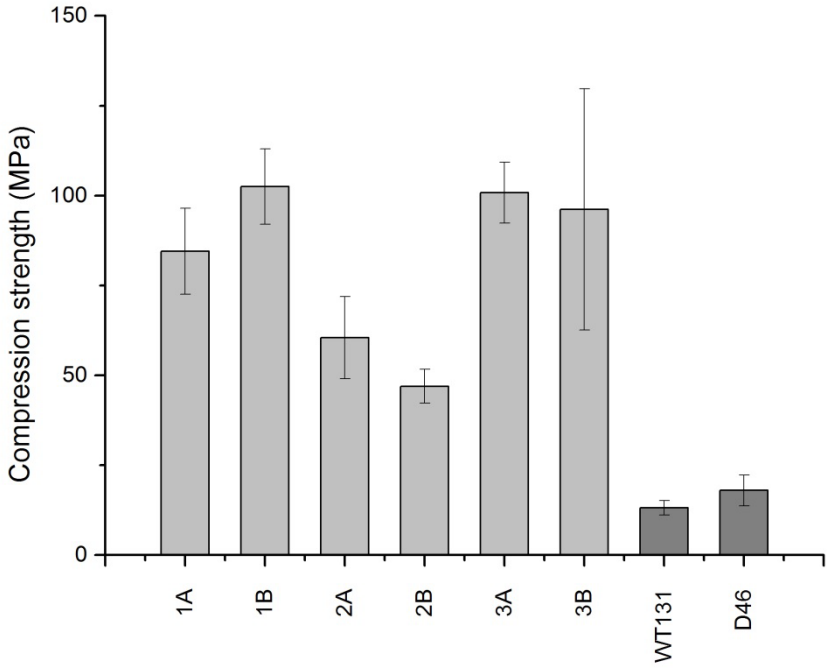


Figure 4. The compression strength of the six tested compositions set against the compression strengths of two regular red bricks (WT131 and D46). D46 was fired at the same brickworks and under similar conditions as 1A, 2A and 3A.

The adsorption curves are shown in Figure 5. The moisture content (U) is defined according to ISO 12571 (2000) as $(m_1-m_0)/(m_0)\cdot 100\%$, where m_0 is the initial dry mass of the tested square prism and m_1 is the mass of the same prism when equilibrium with the environment is reached. All samples adsorbed moisture at very high relative humidity and only up to a few percent. As these determinations are based on the measurements of a single square prism, only the general trend can be outlined from the investigations. For compositions 2B and 3B the adsorption appeared to start at low relative humidity, whereas it was only apparent at > 80 % relative humidity for compositions 1A, 1B, 2A and 3A.

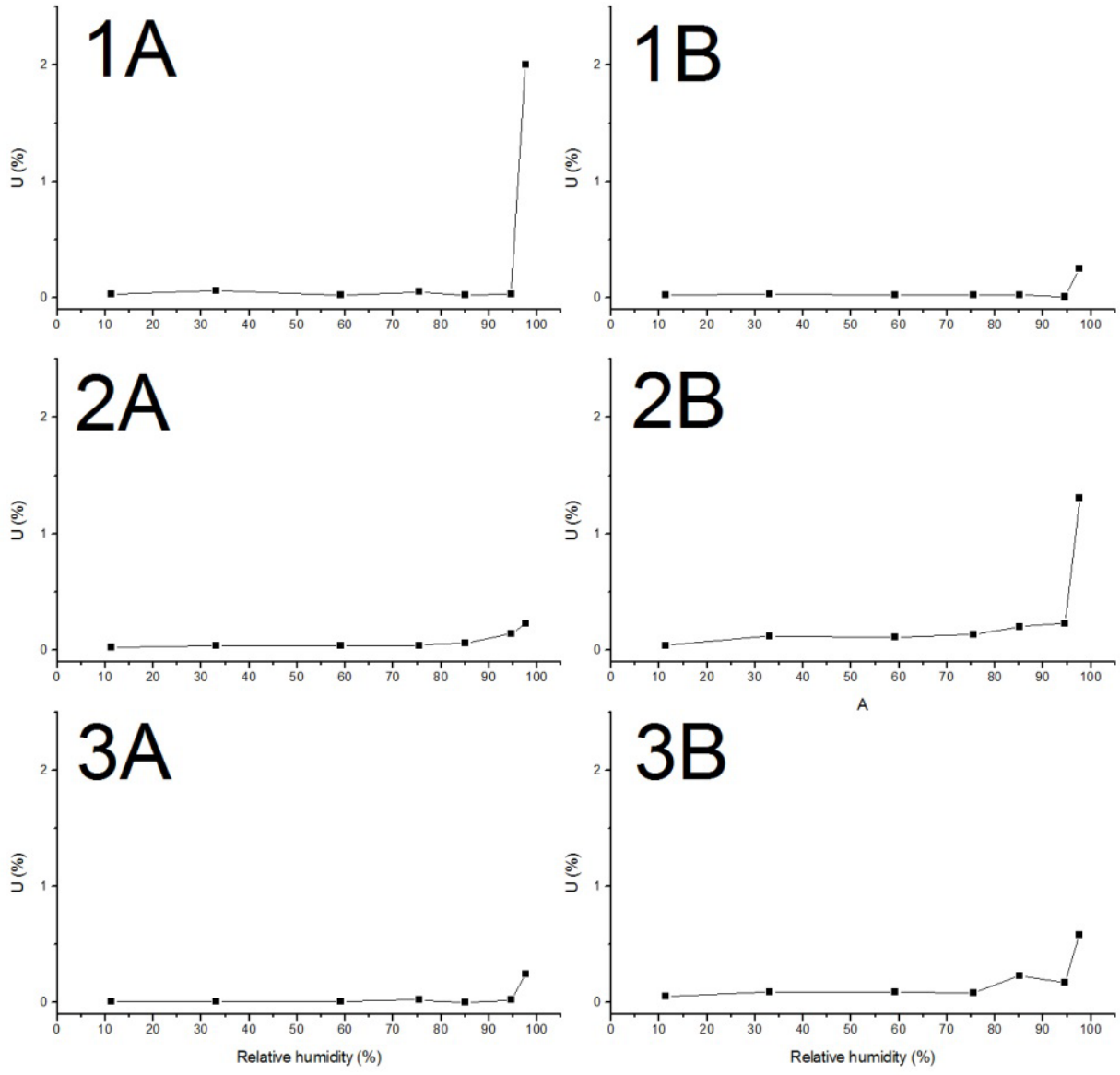


Figure 5. The adsorption curves of the six tested compositions measured at 20°C ± 2°C. Each point on the curves represent a single sample.

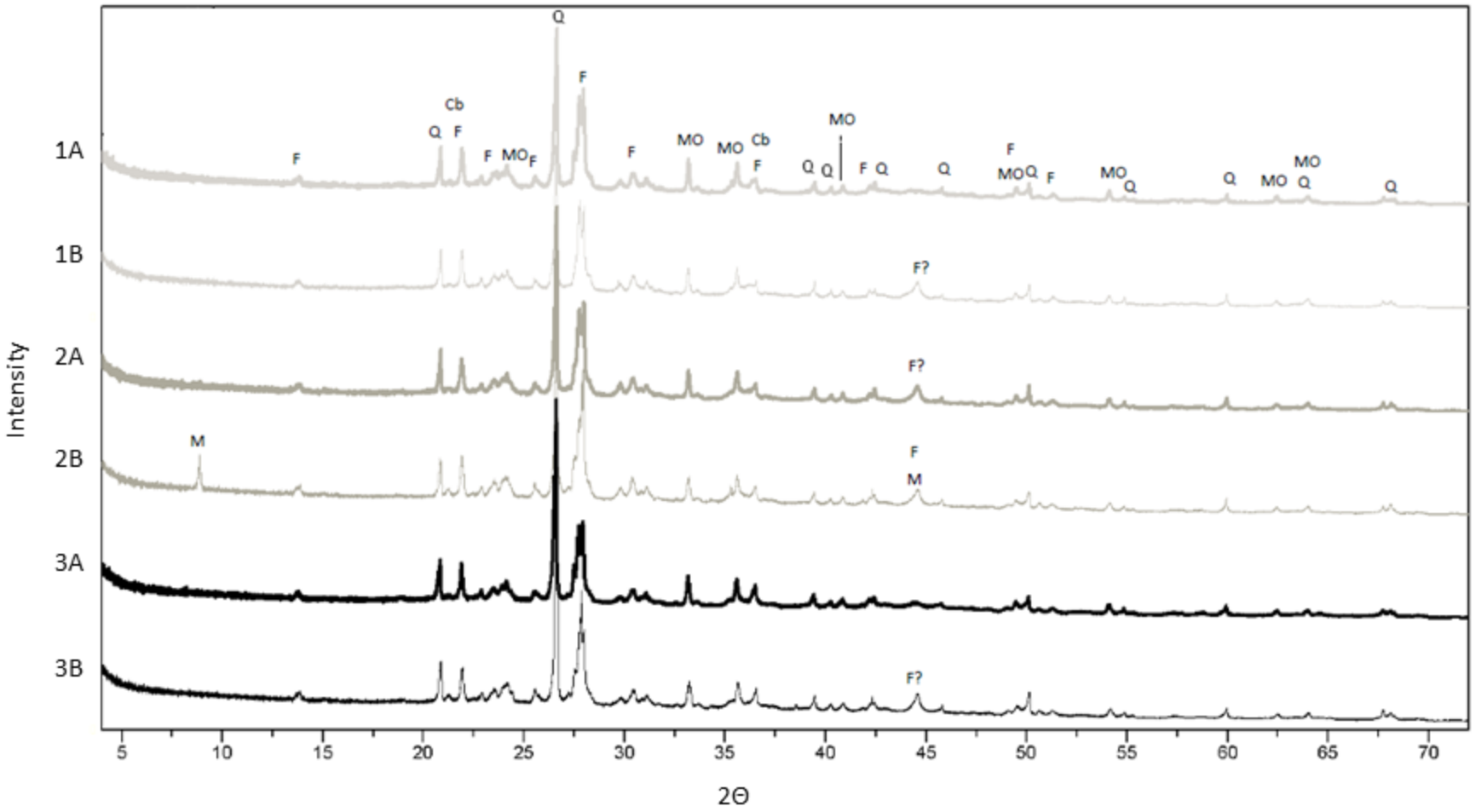


Figure 6: X-ray diffractograms of the fired specimens. The phases identified for composition 1A apply to all specimens and exceptions are noted on the individual patterns. The identified phases are: Cristobalite (Cb), feldspar (F), Mica (M), Metal oxide (MO) and quartz (Q).

The evaluation of the preliminary freeze-thaw test did not demonstrate any failures of the tested bricks (results not shown here). The results from the bulk mineralogy analyses of all the fired compositions are shown in Figure 6. All patterns had inherited features from MS (compared with Figure 3A). Most of the fired specimens lacked phases such as chlorite, mica and amphibole in comparison to MS, but had gained metal-oxide (hematite) and showed indications of the high temperature phase, cristobalite. Peak shifts were observed for some of the feldspar peaks and were likely explained by high temperature phase transitions, e.g. microcline to sanidine.

4. Discussion

Raw material characterisation of MS

The particle size distribution, mineralogy, major element distribution, carbon content and Atterberg limits showed that MS is very similar to other known marine clays from Greenland, Canada and Norway (Belmonte et al., 2014; Bentley and Smalley, 1978; Gillott, 1971; Gillott, 1979; Locat et al., 1984; Pederstad and Jørgensen, 1985; Ramesh and d'Angeljan, 1995; Roaldset, 1972). The relatively low plasticity index of MS presents a challenge when producing bricks by the soft mud method. The extrusion (stiff mud) or dry-pressing methods (Brick Industry Association, 2006) would therefore possibly be better for industrial production using this type of clay.

The choice of compositions and firing temperature

The pure MS compositions, 1A and 1B were fired initially (at 1045-1055°C and 1070-1080°C, respectively), in order to observe the effect of the different temperatures on the technical properties of the produced bricks. The higher firing temperature of 1B led to an increase in total shrinkage, bulk density and compression strength and a decrease in water absorption and open porosity. However, no obvious benefits, which could justify the higher temperature and therefore energy consumption, was discovered for 1B, and it was therefore decided to use lower firing

temperatures of 1030-1055°C for compositions 2A, 2B, 3A and 3B. Furthermore, the relatively high total shrinkage of both 1A and 1B, gave rise to the idea of introducing the two additives, CR and CH, both of which had a coarser grain size distribution and a lower LOI than MS and therefore were less likely to contribute to shrinkage.

Total shrinkage

As expected the total shrinkage is reduced for the CR-containing composition (2A) in comparison to the pure MS composition (1A). However, in spite of a coarser grain size distribution of CH compared to MS, there is no observable difference in total shrinkage between compositions 3A and 1A. Lowering the firing temperature to 1030-1040°C for compositions 2B and 3B also reduced the total shrinkage. According to the Brick Industry Association (2006) the total size shrinkage after drying and firing can vary for different clays, but will usually be around 4.5-8%. For the compositions tested here, only 2B fall within the higher end of this range. However, although the total shrinkage is generally between 11-15% for the compositions in this study, it does not appear to be detrimental, as only very few cracks were observed in the surfaces of the specimens after firing. Most of these cracks were correlated to underlying larger rock fragments and could possibly have been avoided by crushing or by particle-size sorting of the material prior to production, which is standard practice in industrial brick manufacturing (Brick Industry Association, 2006; Hendry and Khalaf, 2001).

Initial rate of water absorption and hygroscopic behaviour

According to ASTM C216-14 (2014) the initial rate of water absorption (IRA) for facing bricks should have a value of up to 1.5 kg/(m^2·min). Bricks with higher IRA values require pre-wetting, as they will otherwise prevent the complete hydration of the cement, due to absorption of water from mortar. Bricks with low IRA should be covered on the jobsite in order to prevent wetting, as

this might prevent or delay the formation of bonds between the brick and the cement. The compositions 1A, 1B, 2A and 3A, which were fired at temperatures of 1045-1080°C, all have low IRA values, whereas the compositions 2B and 3B, which were fired at 1030-1040°C, have IRA values closer to 1.5. All compositions had very low hygroscopic adsorption, however, the adsorption of water at lower relative moisture contents for compositions 2B and 3B compared to the other compositions, indicate that 2B and 3B have larger available inner surface areas where water can be adsorbed. Furthermore, the combined studies on hygroscopic adsorption and capillary uptake indicate that the higher firing temperatures of compositions 1A, 1B, 2A and 3A has reduced the amount of pores capable of capillary uptake compared to the lower temperature compositions 2B and 3B.

Degree of sintering

Vieira and Monteiro (2005) investigated a fine-grained granitic waste material for incorporation in ceramics, which had a d_{50} of approximately 10 µm. They concluded that the fine particle size distribution favoured sintering and reduced the porosity in the produced ceramics. Furthermore the material was shown to have a considerable fluxing potential at temperatures around or slightly above 1000°C, due to a high amount of alkaline oxides present in the form of biotite and Na,K-feldspar, which contributed to the formation of a liquid phase. As both the grain size distribution and qualitative mineralogy of the material characterised by Vieira and Monteiro (2005) are comparable to MS, it is likely that similar reactions would occur here. In addition, the powdery texture (i.e. very fine particle sizes) observed on the surfaces of the grains in MS, is also likely to enhance sintering. When comparing the XRD results of the raw materials to the XRD results of the fired compositions, it is evident that, phases such as mica (biotite), amphibole and chlorite have disappeared after firing, which could be explained by break down/melting of these phases at or below the temperatures investigated in this study. Interestingly, composition 2B, which was fired

at 1030-1040°C still contains some mica (peak at approximately 8.9°2Θ), which is not evident in composition 2A fired at 1045-1055°C. This indicates that at least some of the mica encountered in CR is stable to temperatures above 1030-1040°C but will break down at or below temperatures of 1045-1055°C. The mica in MS appears to break down at temperatures below 1030-1040°C as no mica peaks are apparent from composition 3B, which was fired at this temperature.

The coarser particle size distribution of CR, which has a d_{50} of approximately 600 µm, is less likely to enhance sintering and reduce porosity, which is consistent with the reduced strength and higher open porosity values found for composition 2A compared to 1A.

In general, the strength, density and shrinkage of most of the tested compositions seem to indicate a high degree of sintering of the tested bricks. Future studies should focus on optimising the firing temperature, as it is possible that good quality bricks can still be produced at lower temperatures than investigated in this study.

Durability

A high strength, low water absorption and low open porosity is usually accepted as an indication of high durability of a brick (ASTM C62-13a, 2013; Litvan, 1980; Taylor, 1991), which would also include high resistance to cyclic freezing and thawing. In this study, the investigated properties indicate that the bricks are durable and the preliminary freeze-thaw test did not demonstrate any failures of the tested bricks. This suggests that the bricks could be highly suitable even in climates with high precipitation and regular exposure to cyclic freezing and thawing, such as encountered in the low arctic climate on the West coast of Greenland (Statistics Greenland, 2012) from where the clay originates.

Salt evaporation beneath the surface of a brick, can lead to spalling, and raw materials with large concentrations of salts are therefore avoided or treated prior to production (Mezencevova et al., 2012; Hamer and Karius, 2002; Taylor, 1991). The tested specimens in this study were carefully

examined after the completion of the capillary and porosity measurements (where they had been wetted and subsequently dried), however, no efflorescence of salts was observed. This is consistent with the low chloride concentration ratio, which indicates that although MS had a marine origin, the original salt content in the pore water has since been leached out.

5. Conclusion

The results presented in this paper demonstrate that the marine sediment from Ilulissat (MS) is suitable as brick clay and that it is possible to produce high quality bricks, which can be used in the arctic climate. The produced specimens were fired at temperatures between 1030-1080°C. The highest temperatures did not contribute to further enhancements of the technical properties and the temperature range of 1030-1055°C was therefore chosen for further experiments. The specimens fired at 1045-1055°C generally had very high compression strengths, low porosities and low water absorptions. The introduction of 10 % of the crushing residue, CR, gave rise to a reduction in strengths, higher porosities and water absorptions. In addition, the specimens fired at 1030-1040°C had the lowest strengths, highest porosities and water absorptions. The preliminary freeze-thaw cycles revealed no signs of failure. Due to the similarities in mineralogy, chemistry, plasticity and grain size of MS and other marine sediments found in former glaciated areas of the northern hemisphere, it is likely that the results obtained in this paper are applicable to wide-spread occurrences of similar type of sediments throughout the northern hemisphere.

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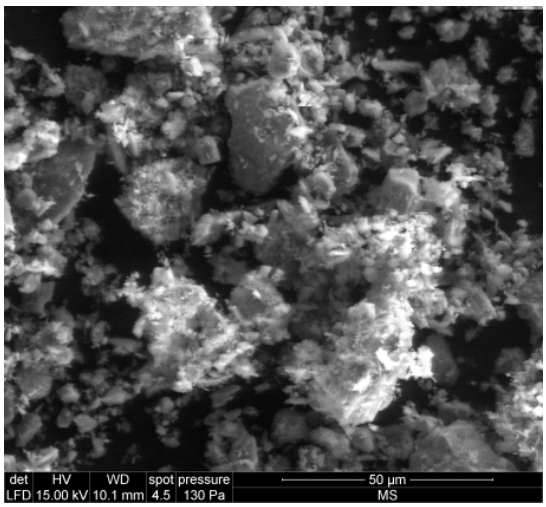
Supplementary material to Appendix 3

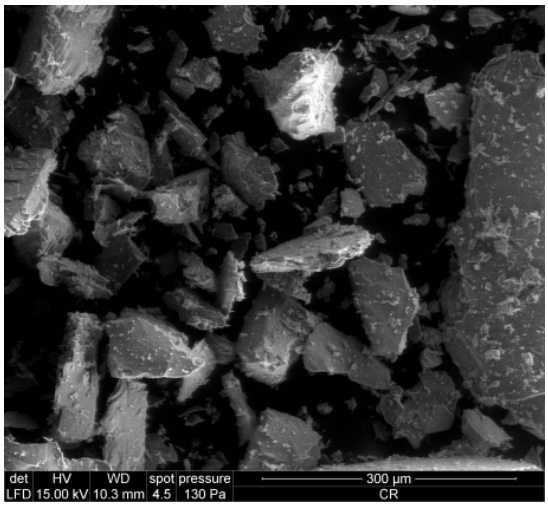
Description: 1) SEM images of MS, CR and CH

2) Photos of the test bricks made from the Ilulissat marine sediment

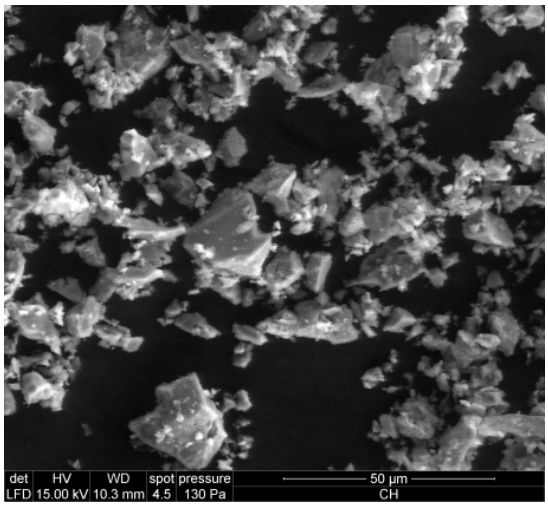
MS



CR



CH



SEM images of MS, CR and CH. Notice the larger scale of magnitude for CR.

Composition: 1A

Specimen name: I1K

Description: 100 % Ilulissat marine sediment, coal-fired at 1045-1055°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing

Dimensions: L: 16.3 cm W: 10.3 cm H: 4.9 cm

Composition: 1A

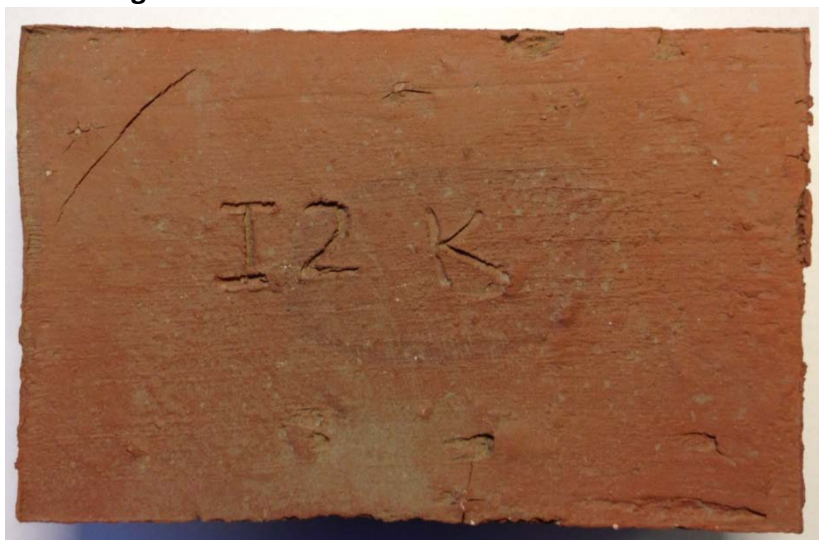
Specimen name: I2K

Description: 100 % Ilulissat marine sediment, coal-fired at 1045-1055°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing



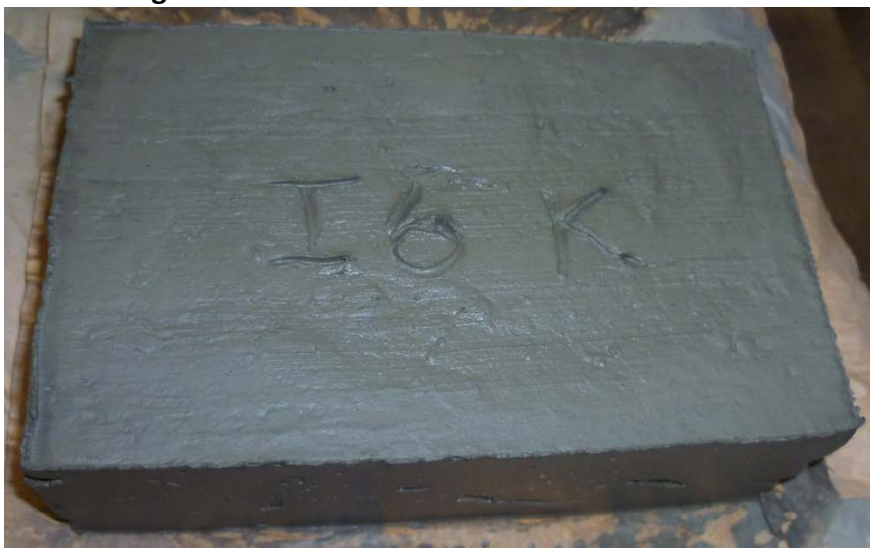
Dimensions: L: 16.4 cm W: 10.4 cm H: 4.6 cm

Composition: 1A

Specimen name: I6K

Description: 100 % Ilulissat marine sediment, coal-fired at 1045-1055°C

Before firing



Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing

Dimensions: L: 16.3 cm W: 10.5 cm H: 4.9 cm

Composition: 1B

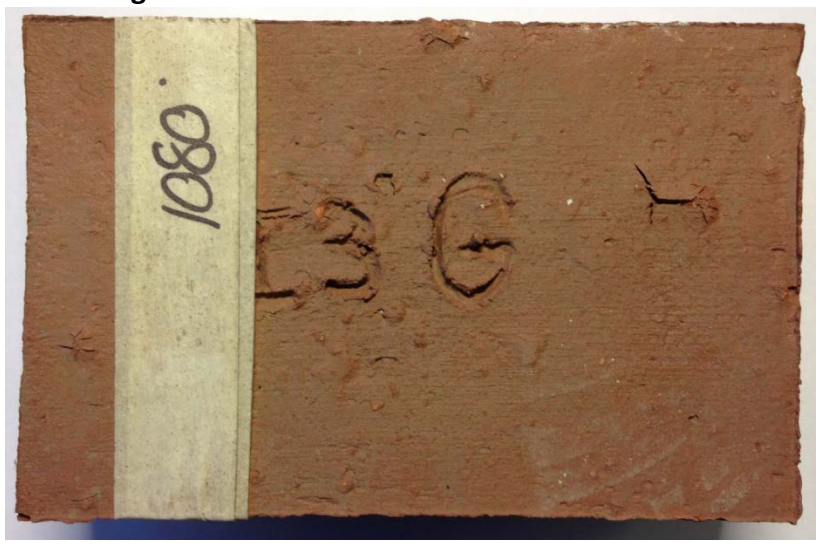
Specimen name: I3G

Description: 100 % Ilulissat marine sediment, fired at 1070-1080°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing



Dimensions: L: 16.1 cm W: 10.3 cm H: 5 cm

Composition: 1B

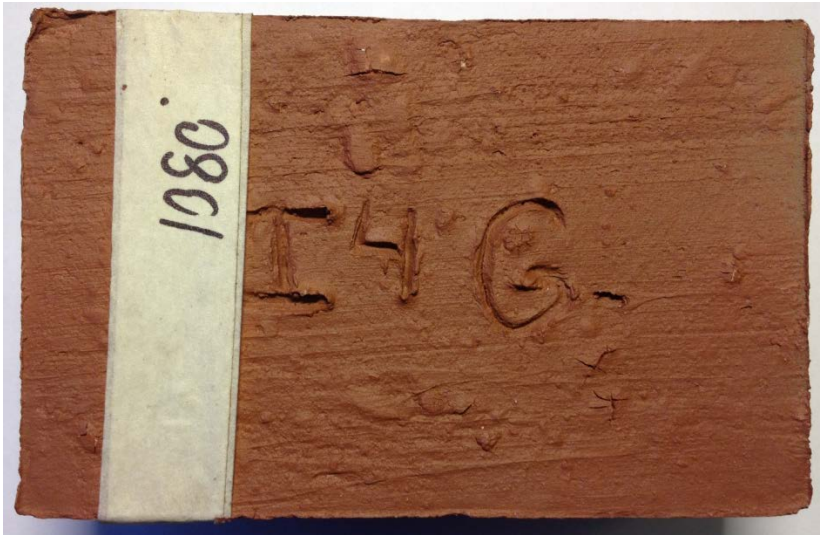
Specimen name: I4G

Description: 100 % Ilulissat marine sediment, fired at 1070-1080°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing



Dimensions: L: 16.0 cm W: 10.2 cm H: 5 cm

Composition: 1B

Specimen name: I5G

Description: 100 % Ilulissat marine sediment, fired at 1070-1080°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing

Dimensions: L: 16.2 cm W: 10.3 cm H: 4.8 cm

Composition: 2A

Specimen name: I7K

Description: 90 % Ilulissat marine sediment + 10% crushing residue (CR), coal-fired at 1045-1055°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing

Dimensions: L: 16.8 cm W: 10.8 cm H: 5 cm

Composition: 2A

Specimen name: I8K

Description: 90 % Ilulissat marine sediment + 10% crushing residue (CR), coal-fired at 1045-1055°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing

Dimensions: L: 16.9 cm W: 10.8 cm H: 5 cm

Composition: 2A

Specimen name: I9K

Description: 90 % Ilulissat marine sediment + 10% crushing residue (CR), coal-fired at 1045-1055°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing

Dimensions: L: 16.7 cm W: 10.8 cm H: 5.1 cm

Composition: 2B

Specimen name: I10G

Description: 90 % Ilulissat marine sediment + 10% crushing residue (CR), fired at 1030-1040°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing

Dimensions: L: 17.3 cm W: 11.3 cm H: 5.3 cm

Composition: 2B

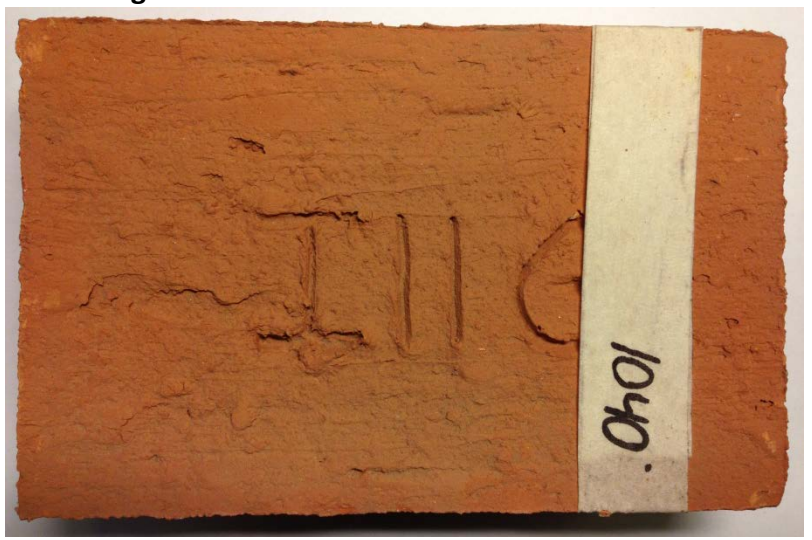
Specimen name: I11G

Description: 90 % Ilulissat marine sediment + 10% crushing residue (CR), fired at 1030-1040°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing



Dimensions: L: 17.6 cm W: 11.4 cm H: 5.2 cm

Composition: 3A

Specimen name: I16K

Description: 90 % Ilulissat marine sediment + 10% chamotte (CH), coal-fired at 1045-1055°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing

Dimensions: L: 16.3 cm W: 10.4 cm H: 4.9 cm

Composition: 3A

Specimen name: I17K

Description: 90 % Ilulissat marine sediment + 10% chamotte (CH), coal-fired at 1045-1055°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing

Dimensions: L: 16.3 cm W: 10.4 cm H: 5.1 cm

Composition: 3A

Specimen name: I18K

Description: 90 % Ilulissat marine sediment + 10% chamotte (CH), coal-fired at 1045-1055°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing

Dimensions: L: 16.3 cm W: 10.4 cm H: 5.1 cm

Composition: 3B

Specimen name: I13G

Description: 90 % Ilulissat marine sediment + 10% chamotte (CH), fired at 1030-1040°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing

Dimensions: L: 16.8 cm W: 10.8 cm H: 5.3 cm

Composition: 3B

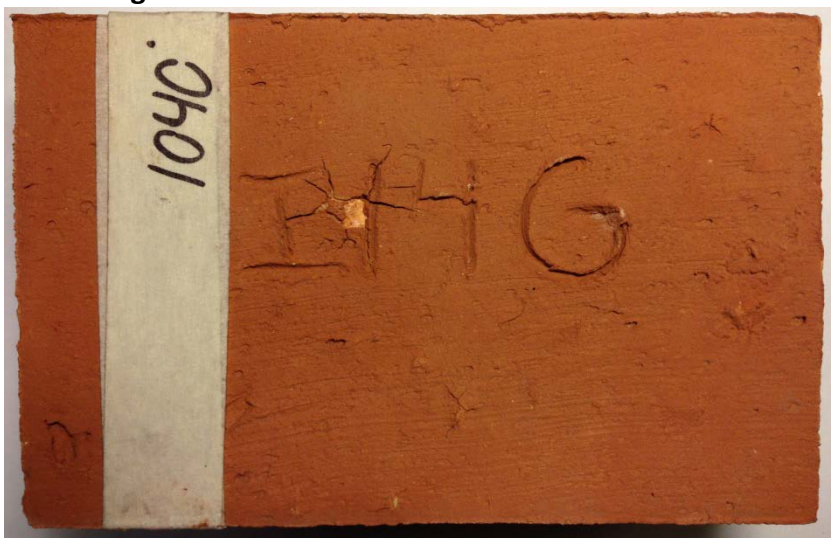
Specimen name: I14G

Description: 90 % Ilulissat marine sediment + 10% chamotte (CH), fired at 1030-1040°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing



Dimensions: L: 16.9 cm W: 10.3 cm H: 5.4 cm

Composition: 3B

Specimen name: I15G

Description: 90 % Ilulissat marine sediment + 10% chamotte (CH), fired at 1030-1040°C

Before firing

Dimensions: L: 18.9 cm W: 12 cm H: 6 cm

After firing

Dimensions: L: 16.8 cm W: 10.8 cm H: 5.3 cm

Appendix 4

Description: Conference paper. Proceedings of the conference: Rock mechanics in the Nordic countries, Kongsberg, Norway, 9-12th of June 2010.

Title: Use of clay from Kangerlussuaq in the Greenlandic construction industry

PROCEEDINGS

Bergmekanikk i Norden 2010

Rock mechanics in the Nordic countries 2010

KONGSBERG, NORWAY 9. – 12. June 2010

Organisert av:

NORSK BERGMEKANIKKGRUPPE

Tekna

Sponsorer:

Statens vegvesen

Norges Geotekniske Institutt
NGI
Norwegian Geotechnical Institute

Norconsult

USE OF CLAY FROM KANGERLUSSUAQ IN THE GREENLANDIC CONSTRUCTION INDUSTRY

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Arctic Technology Centre, BYG•DTU, Technical University of Denmark

SUMMARY

Clay material from Kangerlussuaq in West Greenland was characterised and its possible use for the production of bricks, expanded clay products and inert filler material was investigated. It was generally found that it was possible to use the clay in all of the above mentioned materials, although, further investigations will be needed before a final conclusion can be made. This paper is based on results and conclusions of student projects that were carried out at the Arctic Technology Centre, DTU.

INTRODUCTION

Raw materials for use in the Greenlandic construction industry are shipped to Greenland from all over the world. The materials are expensive to import and therefore reuse and use of resources already in Greenland would make a huge contribution to the local community. At the Arctic Technology Centre several student projects have looked into the possibilities of finding new raw materials in Greenland. In order for these materials to be competitive with imported materials, it is important to consider various aspects such as accessibility, amount, characteristics and possible uses of the new material. One raw material, which has been considered, is clay from near the town of Kangerlussuaq in West Greenland. During the past ten years several student projects have been concerned with the mapping and characterization of the clay and with finding and testing the possible uses in materials for construction, such as bricks, inert filler material and expanding clay products. This paper summarizes some of the results and conclusions of the investigations concerned with the clay from Kangerlussuaq.

GEOGRAPHY AND GEOLOGY

The town of Kangerlussuaq is located in the western end of the 185 km long Kangerlussuaq fjord (see figure 1). The fjord is supplied with fresh water and sediments from the Russel Glacier (located approximately 25 km west of the town) by two major rivers (see figure 1).

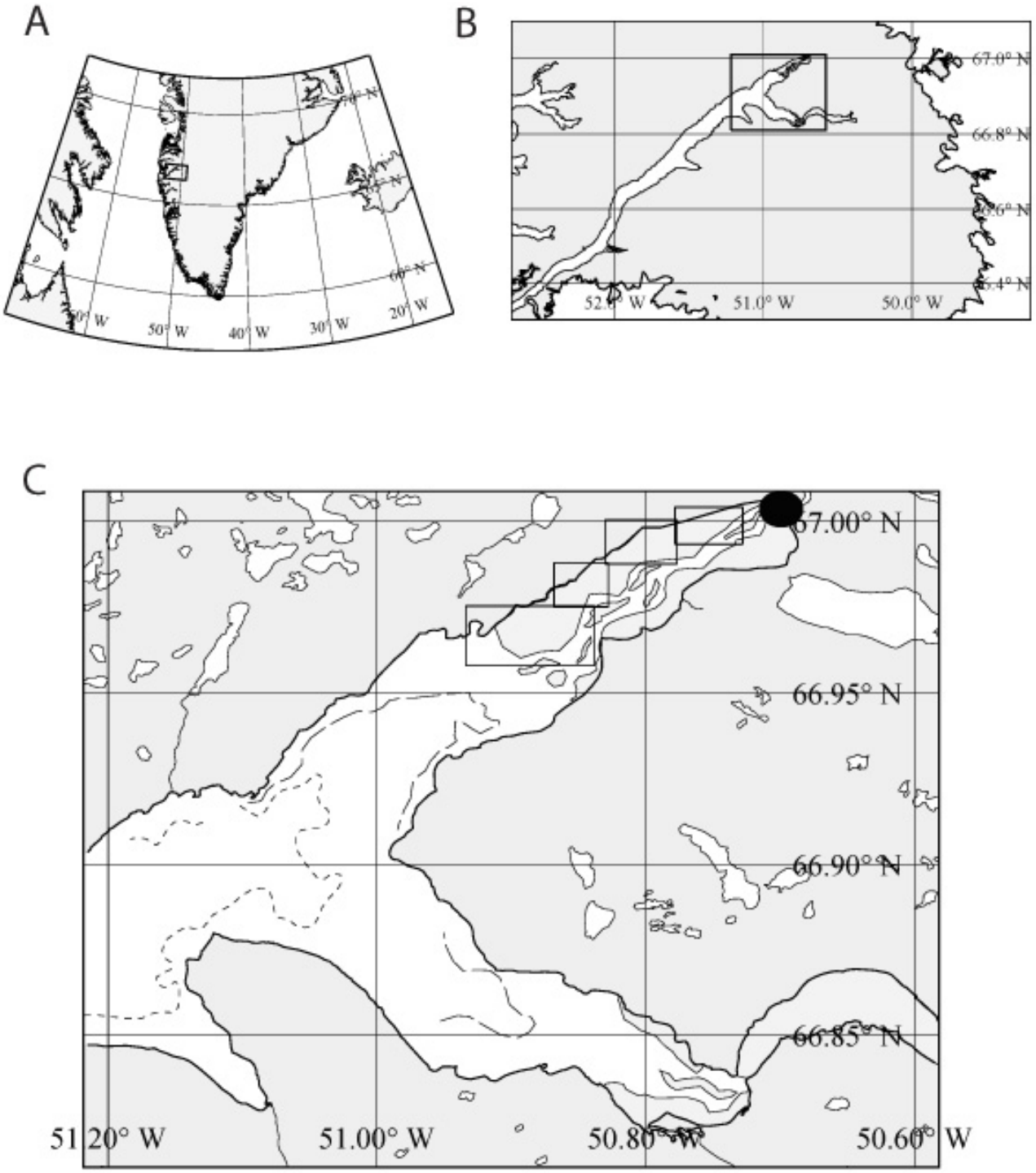

Figure 1. Overview maps of Greenland and the Kangerlussuaq fjord. **A.** Greenland. The black square indicates the location of map B. **B.** A section of the Kangerlussuaq fjord. The icecap (outlined by a black line) which supplies the fjord with fresh water and sediments is located west of the fjord. The black square indicates the location of map C. **C.** The inner part of the Kangerlussuaq fjord where clay formations are common. The town of Kangerlussuaq is marked by a black circle. The clay was sampled from several localities within the smaller black squares.

The sediments are primarily formed by mechanical erosion of the rock underlying the icecap and sorted by transport in the rivers before deposition. Chemical weathering is believed to be negligible due to the cold and arid climate in the inner part of the Kangerlussuaq fjord. The town of Kangerlussuaq is situated on a delta where the glacial stream water meets the saline fjord water,

which gives rise to flocculation and sedimentation. Furthermore, several sediment plains are now exposed above sea level, due to isostatic rebound since the last ice age.

The Kangerlussuaq fjord is situated in the border zone between the Archean Craton to the South and the Nagssugtoqidian mobile belt to the North [1]. The dominating rock types in the area are Archean orthogneisses (granulite-facies), Archean plutonic rocks (granitic to tonalitic) and Proterozoic gneisses (amphibolites-facies). Several ultramafic dykes (kimberlites and lamprophyres) and a carbonatite complex are found in the area and belong to a large regional alkaline province. It is likely that all of the rock types in the area can be found underneath the icecap as well, and therefore leave their mineralogical imprint on the sediments found in the inner part of the Kangerlussuaq fjord.

MATERIAL CHARACTERISATION

The samples described here were primarily collected from a sediment plain, locally known as "Fossilsletten" (see figure 2) and from clay deposits in close vicinity of the town (see figure 1)

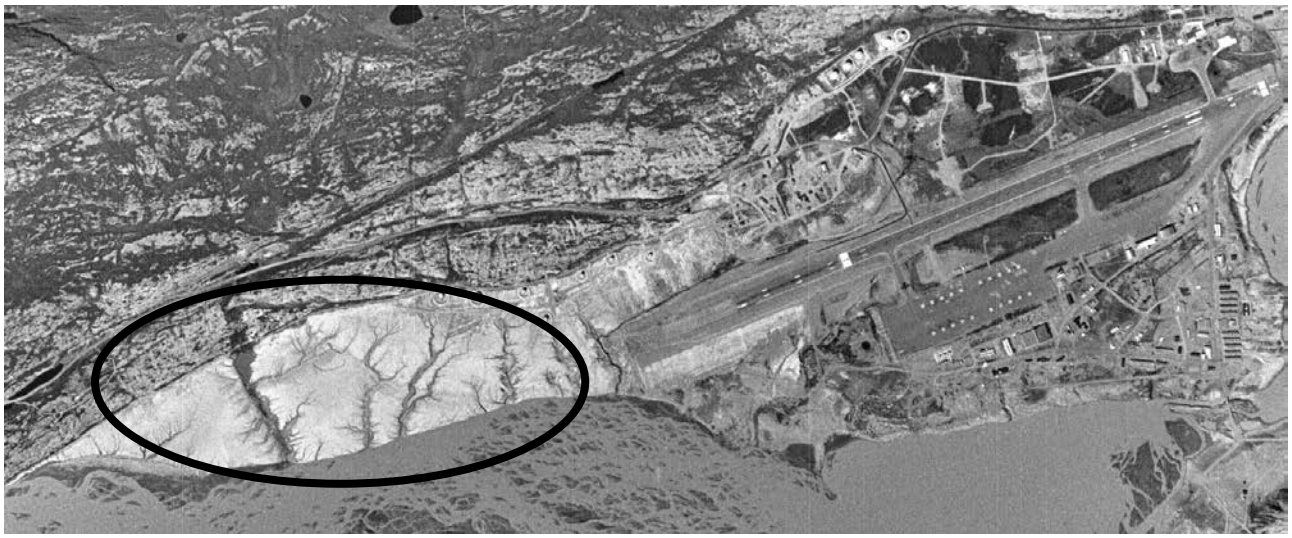

Figure 2. Aerial photograph of the town of Kangerlussuaq from 1986. The sediment plain "Fossilsletten" (marked by the black circle) is located west of the airport.

Grain size distribution

The grain size distribution was measured for samples from different parts and depths of "Fossilsletten" and from the fjord [2, 3, 4, 5]. The grain-size distributions of the samples from "Fossilsletten" are shown in Figure 3. According to Larsen *et al.* (1995) [6] the sediment can generally be described as fine grained with 45-60% in the clay size fraction (< 0.002 mm), 30-45% in the silt fraction (0.002-0.06 mm) and up to 10% sand (0.06-2 mm). Alm & Brix (2005) [5] described a few samples from the fjord that were coarser with a larger representation of the sand fractions (up to 1.3 mm). In general, though, the material from "Fossilsletten" can be considered quite homogeneous in what concerns the grain size distribution, which is important if the material is to be used as a raw material for the production of construction materials.

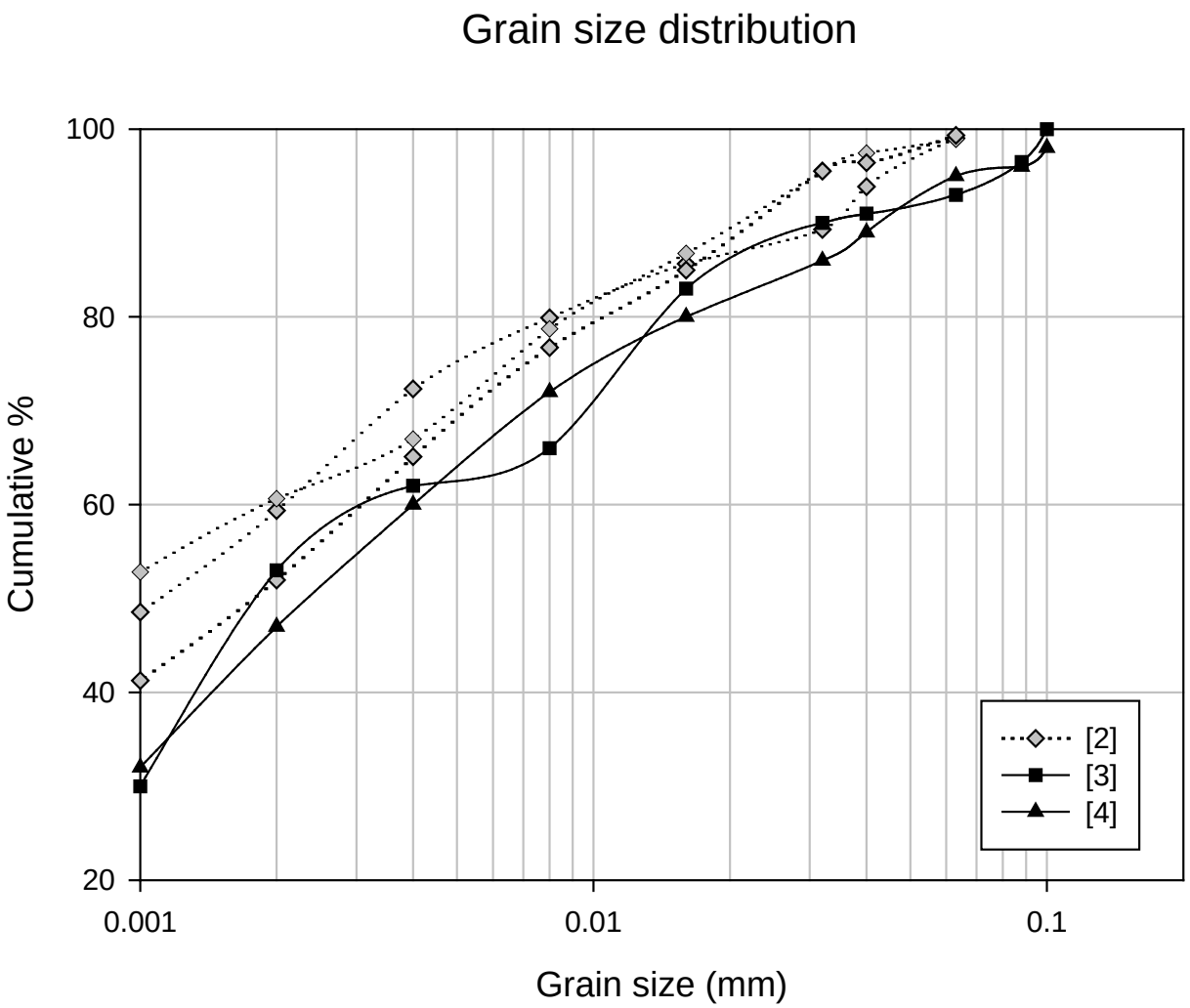

Figure 3. The grain size distribution of the Kangerlussuaq clay as measured by Jacobsen *et al.* (2002) [2], Sørensen & Welinder (2003) [3] and Morelli & Bennedsen (2005) [4]. The samples shown here are from "Fossilsletten" and were obtained from different depths and localities. Samples obtained from the fjord by Alm & Brix (2005) [5] (not shown in this figure) generally show a similar trend, however, a few samples were coarser with grain sizes up to 1.3 mm.

Mineralogy and bulk chemistry

The mineralogy of some of the clay samples from Fossilsletten and the fjord were analysed using X-ray powder diffraction [5, 2]. The samples contained: plagioclase (33.8-44.6 wt%), quartz (14.7-29.4 wt%), pyroxene (10.3-23.7 wt%), amphibole (3.0-16.7 wt%), biotite (0.0-15.1 wt%), K-feldspar (0.0-13.8 wt%) and minor amounts of other minerals such as chlorite. Jacobsen *et al.* (2002) [2] concluded that the mineralogy of several samples from Fossilsletten was almost identical. The mineralogy appears to be consistent with the mineralogy of the dominating rock types in the area.

The bulk chemistry of several samples from Fossilsletten was determined by XRF [2] and gave the following bulk chemistry: 57.5 wt% SiO_2, 0.8 wt% TiO_2, 16.1 wt% Al_2O_3, 2.7 wt% Fe_2O_3, 3.8 wt% FeO, 0.1 wt% Mn_3O_4, 3.8 wt% MgO, 4.4 wt% CaO, 4.5 wt% Na_2O, 2.9 wt% K_2O, 0.2 wt% P_2O_5 and 2.4 wt% volatiles, adding up to an oxide total of 99.2 wt%.

C, $CaCO_3$, Fe and S content

The C, $CaCO_3$, Fe and S content were measured for some of the samples and are listed in table 1. In all studies the C and S contents were determined using a LECO analyzer and the Fe contents were determined using atomic absorption spectrometry after acid digestion. The $CaCO_3$ contents were either determined through titration with HCl (marked with T in table 1) or estimated from the results on C from the LECO-analyzer (marked with L in table 1).

Table 1. List of C, $CaCO_3$, Fe and S analyses. The $CaCO_3$ contents were either determined through titration with HCl (T) or estimated from the results on C from the LECO-analyzer (L).

	[4]	**[2]**	**[7]**	**[3]**
Carbon content	below 0.85 wt%	below 0.06 wt%	below 0.12 wt %	0.04 wt %
$CaCO_3$ content	~ 5 wt% (T)	2.9 wt% (T)	2.6 wt% (T)	max. 0.01 wt % (L)
Sulphur content	below 0.06 wt%	below 0.04 wt%	below 0.06 wt%	0.06 wt %
Fe content	3.23 wt %	3.46 - 3.95 wt%	2.92-3.07%	3.4 wt %

Salt content

The salt content was measured for some of the samples from "Fossilsletten". Jacobsen *et al.* (2002) [2] found that the salt content, measured as the chloride content, decreased from the surface to a depth of approximately 1-1.5 m, where it increased until permafrost was reached at a depth of approximately 2-2.5 m. The salt content was not measured below the depth of permafrost. The content ranged from 1.4-2.3 wt%. Sørensen & Welinder (2003) [3] took several surface samples with a chloride content ranging from 0.7-2.3 wt%, but found no systematic explanation for the variation.

Water content

The water content was measured for some of the samples from "Fossilsletten". According to Jacobsen *et al.* (2002) [2], the water content increases with depth. Due to the arid climate in the inner part of the Kangerlussuaq fjord, the evaporation is larger than the precipitation, and the water content is therefore lower near the surface than at depth. From the surface level to a depth of approximately 0.5 m the water content was measured at different localities and was found to be between 15-20%. From a depth of 0.5-2.5 m the water content was between 20-25%.

Loss on ignition

The Kangerlussuaq clay has a low loss on ignition (below 3%) at temperatures up to 1000°C, which indicates that water-containing minerals, such as the clay minerals are scarce. Furthermore, this indicates a low content of organic material and $CaCO_3$ [2,7].

POTENTIAL USE OF THE KANGERLUSSUAQ CLAY

It was estimated that "Fossilsletten" consists of at least 11.5 million m^3 of clay [2] and, furthermore, additional clay can likely be recovered from the fjord. The extent and accessibility of the Kangerlussuaq clay could make it an interesting local Greenlandic resource for production of construction materials. In the following the investigations on the potential use of the clay is described.

Inert filler material

In Greenland all cement is imported, which makes it an expensive material. In the concrete or mortar production it is possible to replace some of the cement with an inert filler. An inert filler is a fine grained material, that will be able to fill some of the pore spaces between the aggregates and thereby reduce the necessary amount of cement in the mortar/concrete. The study by Alm & Brix (2005) [5] suggests that the grain size and mineralogy of the Kangerlussuaq clay makes it suitable as inert filler material, however, the high chloride content is problematic as it can give rise to corrosion damage in concrete. Alm & Brix (2005) [5] substituted increasing amounts of the clay for Aalborg Portland Rapid cement in mortar samples in order to study the influence on the strength of the cement. They concluded that up to 10% of the cement could be replaced by the Kangerlussuaq clay material; however, further analyses on especially the effect of the chloride content will be needed before a final conclusion can be reached.

Bricks

Bricks can be made by moulding, drying and firing/burning clay and they are commonly used for building constructions. The Fe and $CaCO_3$ content in the clay have a big influence on the colour of the finished brick. When the mass of $CaCO_3$ is at least 3 times higher than the mass of Fe_2O_3 the bricks become yellow. A lower mass ratio of $CaCO_3$ to Fe_2O_3 will give red colours. Jacobsen *et al.* (2002) [2] found that bricks made from the Kangerlussuaq clay gained a pale-red colour. Jacobsen *et al.* (2002) [2] argued that the Kangerlussuaq clay was suitable for bricks, apart from the very high salt content. The high salt content could be problematic for some types of furnaces and it could also lead to salt efflorescence on the surface of the finished bricks. However, further investigations would be needed to confirm whether or not the salt would pose a serious problem for brick production. In such case the salt could be washed out of the clay by water prior to use.

Expanding clay products

Expanding clay products are formed by partial melting at temperatures above 1100°C. The clay material used should be able to develop a highly viscous melt and at the same time develop gasses. The gasses which are trapped by the melt will then expand the product. Expanding clay products can, for example, be used as insulation in buildings or as filler material in roads. Based on chemical analyses, Morelli & Bennedsen (2005) [4] found that the Kangerlussuaq clay lacked $CaCO_3$ and C to enhance the melting and gas formation. Villumsen & Frydenlund (2006) [8] demonstrated that $CaCO_3$ and C containing crab waste from Greenlandic factories could be used as an additive to the clay and a patent was granted for the invention of expanding clays using crab waste [8]. Based on experiments, Morelli & Bennedsen (2005) [4] concluded that it was possible to use the Kangerlussuaq clay for expanded clay products, although further experiments would be needed in order to optimize the composition and conditions for the production. Morelli & Bennedsen (2005) [4] estimated that the high salt concentration in the clay could pose a problem for the furnaces in a large scale production; furthermore, it was believed that a higher Na-content in the clay would be beneficial and that there might be other more suitable clay formations for expanding clay products in Greenland.

Possible use of bricks and expanded clay products in Greenland

Løjmand & Overgaard (2007) [9] investigated whether Greenlandic made bricks and expanded clay products would be suitable materials for structures in the arctic climate and, furthermore, whether their use would be profitable. Up to now, bricks and expanded clay have not been widely used in the Greenlandic construction industry, where wood and concrete have been used predominantly; however, many housing estates in Greenland that were built during the 1960s are now in a poor condition and will need to be replaced or renovated within a few years, which will raise the demand for suitable construction materials. Løjmand & Overgaard (2007) [9] evaluated the physical and technical properties of bricks and expanded clay products in general and concluded that the materials would be suitable construction materials in the arctic climate. Furthermore, preliminary investigations suggested that there could be an economic benefit in using these materials, however, further investigations would be needed in order to evaluate whether a Greenlandic production of the materials would be profitable.

Conclusions

“Fossilsletten” consists of at least 11.5 million m^3 of relatively homogeneous clay and could be an interesting local Greenlandic resource for the production of construction materials. Preliminary investigations have shown that the clay can be used as inert filler material in mortar or concrete production, and for the production of bricks and expanded clay products. There is still research to be conducted to fully evaluate the potential for utilizing the clay resource, and the investigations showed that there are several possibilities which should be explored further.

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9. Løjmand, T. and Overgaard, A.S. ”Tegl og letklinker anvendt i grønlandsk byggeri”. Unpublished report, Arctic Technology Centre, BYG•DTU, 2007

Appendix 5

Description: Unpublished paper

Title: Screening of Greenlandic MSWI ashes and mine tailings for use in clay-based bricks

Screening of Greenlandic MSWI ashes and mine tailings for use in clay-based bricks

Belmonte, L. J.[a,*], Ottosen, L. M.[a], Kirkelund, G. M.[a], Jensen, P. E.[a] and Vestbø, A. P.[b]

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Abstract

Municipal solid waste incineration (MSWI) fly and bottom ashes and mine tailings (i.e. residues from the mineral resource industry) from Greenland were screened in order to determine their suitability as secondary resources in clay-based brick production. The screening involved characterisation of the materials, production of small scale brick pellets (discs with a diameter of approximately 2 cm) and determination of properties such as density, porosity and water absorption. Heavy metal leaching tests were conducted before and after firing. The mine tailings and bottom ash brick pellets obtained satisfactory densities (1669-2007 kg/m^3) and open porosities (27.9-39.9 %). In contrast, the fly ash brick pellets had low densities (1313-1578 kg/m^3) and high open porosities (42.1-51.5 %). Remediation techniques (washing in distilled water and electrodialytical treatment) applied to the fly ash reduced leaching of the samples before firing. However, leaching tests on crushed brick pellets revealed that heavy metals generally became more available after firing for all the investigated materials and that initial or further remediation therefore could be necessary prior to incorporation in bricks.

Keywords: MSWI fly ash, MSWI bottom ash, mine tailings, clay-based ceramics, Greenland

1. Introduction

Today, most of the materials for use in the Greenlandic construction industry are imported from Denmark and Europe. Bricks are not currently produced locally nor frequently applied for construction purposes. However, Belmonte et al. (2014, a) and Belmonte et al. (2014, b) established that marine clays from former glaciated areas of the northern hemisphere were suitable materials for the production of bricks. Furthermore, due to the excellent durability, low maintenance, high fire-resistance and good thermal and acoustic properties of bricks in general (Hornbostel, 1991) they were concluded to be a highly suitable construction material in the Arctic (Belmonte et al., 2014, b).

Fired clay bricks, are heterogeneous materials with wide-ranging compositions, which can tolerate the incorporation of waste materials, even in high percentages, without impairing their overall technical properties (Dondi et al., 1997(a,b); Segadães et al., 2005). Utilisation of any waste material would in addition, which at the same time would solve the problem of their disposal (Zhang, 2013). Furthermore, several other advantages, such as inertisation of hazardous substances, better environmental protection, enhanced technical properties and reduced production costs have been mentioned in the literature (Rambaldi et al., 2010; Roy et al., 2007; Torres et al., 2009; Zhang et al., 2007).

The main residues produced by modern MSWI plants are bottom ash (slag) and flue gas cleaning residues such as fly ash (Hjelmar et al., 2011). Bottom ashes are heterogeneous mixtures of ash, metals and other non-combustible materials such as ceramics and glass and typically have grain sizes in the range from 0.06 - 60 mm (Hjelmar et al., 2011). Fly ashes consist of the fine particulate matter, which are entrained in the flue gas and typically have grain sizes in the range of 0.002-2 mm (Hjelmar et al., 2011). The chemical compositions of MSWI bottom and fly ashes vary and depend on the materials being incinerated and the type of incinerator. The ashes can contain

hazardous substances defined according to the European parliament and the council of the European Union (2008) and their use as secondary raw materials might therefore be excluded or restricted according to national legislation. Several researchers have, however, investigated and successfully incorporated MSWI ashes in clay-based ceramics (e.g. Kasuriya et al., 2008; Rambaldi et al., 2010; Zhang et al., 2007; Zhang et al., 2011). In the mentioned studies, the ashes replaced between 2-60 % (by weight) of clay or sand and properties such as reduced leaching of heavy metals after firing (Rambaldi et al., 2010; Zhang et al., 2007 and Zhang et. al., 2011) and enhanced densification (Kasuriya et al., 2008) were reported for the produced ceramics. In Greenland, municipal solid waste management was not a major priority before the 1990's, due to the scarce and geographically wide-spread population. The waste was traditionally handled by uncontrolled incineration and/or dumping at open disposal sites (Eisted and Christensen, 2011; Kirkelund et al., 2012). Increased awareness of the environmental impact has, however, since led to the introduction of a waste strategy and several small municipal solid waste incineration (MSWI) plants. The incineration plants have simple flue gas cleaning systems by which fly ashes are collected in bag- or electrostatic filters. The fly ashes are subsequently exported for deposition in Langøya in Norway while bottom ashes are still dumped at local disposal sites. Alternative handling of the fly ashes is of particular importance, as the export is very costly for the Greenlandic society.

Mine tailings worldwide often present major economic and environmental challenges to both the mining companies and the local community (e.g. Lorber and Antrekowitsch, 2011; Reid et al., 2009; Roy et al., 2007). They are traditionally disposed in large subaerial (or underground) facilities or in subaqueous environments, e.g. lakes or marine water. Due to serious environmental hazards, such as heavy metal pollution, the tailings disposals often need continuous surveillance even after the mining operations have terminated (e.g. Elberling et al., 2002). Several authors have

investigated the potential for using residues from the mineral resource industry as secondary raw materials for the production of clay-based ceramics (e.g. Chen et al., 2011; Menezes, 2005; Roy et al., 2007; Segadães et al., 2005; Torres et al., 2009). Benefits such as increased durability (Torres et al., 2009), reduced firing temperature (Segadães et al., 2005) and lowered total cost (Roy et al., 2007) were reported for the ceramics produced in the above mentioned studies. Up until this date, only a few mines have been in operation in Greenland, but severe environmental problems relating to mine tailings from some of these mines have already been experienced (Johansen and Asmund, 2001).

In this paper, a screening of MSWI ashes and mine tailings from Greenland was conducted in order to determine their potential suitability for incorporation in the production of clay-based bricks. Furthermore, the MSWI fly ash was subjected to two remediation techniques (electrodialytic treatment and washing) with the purpose of studying the effects of these treatments on the leaching behaviour.

2. Materials and methods

2.1 Raw materials

Marine clay

Marine glaciogene clays are known from numerous localities throughout the Arctic and former glaciated areas of the Northern hemisphere (Belmonte et al., 2014, a; Gillott, 1979; Locat et al., 1984; Ramesh and d'Angeljan, 1995; Roaldset, 1972; Rosenqvist, 1975) Approximately 150 kg of marine clay from the town of Ilulissat, West Greenland was sampled from a depth interval of 20-100 cm below surface. The material was roughly homogenised using a large shovel and stored in closed containers at room temperature until use.

Incineration residues

Municipal solid waste incineration (MSWI) plants exist in the six largest towns in Greenland (Kirkelund et al., 2013; Eisted and Christensen, 2011). The combustible fractions from residential waste, commercial waste and construction waste are incinerated and the bottom ash is landfilled at disposal sites without control of leachate and gas (Eisted and Christensen, 2011). Fly ash, which is recovered after treating the flue gas in electrostatic filters, is temporarily stored at the disposal sites before being exported as hazardous waste for further treatment in Denmark. The current annual production of bottom ash and fly ash in Greenland is estimated to be 6000 tonnes and 200 tonnes, respectively (Eisted and Christensen, 2011). The samples used in this study were collected from the MSWI plant in the town of Sisimiut, West Greenland. Both the bottom ash and fly ash were collected as fresh samples from the incineration plant and stored at room temperature until use.

Mine tailings

Mine tailings from two South Greenlandic sites, the Nalunaq Gold Mine and the Tanbreez Mining Greenland A/S project, were investigated in this project.

The Nalunaq Gold Mine is currently not in operation but was at the time of sampling (summer 2012) operated by Angel Mining Plc. The Nalunaq gold mineralisation is classified as a mesothermal vein-type gold deposit hosted in metavolcanic rocks (Secher et al., 2008) and the surrounding region is characterised as a gold province with several promising occurrences (Stendal and Secher, 2002; Steenfelt, 2000). The ore, which consists of a gold-bearing quartz vein (known as the main vein) and proximal hydrothermally altered dolerites and amphibolites (Schlatter and Olsen, 2011), was crushed, milled and treated by cyanide leaching in an extraction plant on-site. The tailings slurry was subjected to detoxification using the Inco method (Lewis, 1984) and subsequently pumped into a tailings dam located in previously exploited levels of the mine. At the

time of sampling, the estimated tailings production was 50,000-70,000 tonnes per year. In this study, tailings were obtained from the discharge of the detoxification tank before going into the tailings dam. The sampled slurry contained 20-22% solids, which were left to settle before the excess water was decanted.

The Tanbreez Mining Greenland A/S project currently holds an exploration licence in the Ilimaussaq intrusion complex. The project focuses on a laminated karkortokite, which consists of separate layers of feldspar, eudialyte and mafic minerals such as arfvedsonite and aegirine (Sørensen, 2001). The on-site ore processing will involve dry crushing followed by magnetic separation. The magnetic separation will produce three concentrates, an eudialyte, a feldspar and a mafic mineral concentrate. Furthermore, a fine-grained residue, representing bulk rock, will also originate from the ore crushing. The mafic mineral concentrate and fine grained residue are currently considered as tailings, which are planned to be disposed on-site. It is expected that the production will result in 230,000 tonnes of tailings per year, whereof the mafic concentrate will constitute approximately 180,000 tonnes and the fine grained residue will constitute 50,000 tonnes. Here, both the mafic concentrate (Tanbreez mafic) and the fine-grained bulk residue (Tanbreez fines) were used.

2.2 Methods

2.2.1 Initial treatments of the raw materials

Large fragments (> 1 mm) in both the marine clay and the bottom ash were removed before further use, by wet and dry sieving respectively. The fly ash was subjected to two different remediation treatments: 1) a portion was washed with distilled water in a 45 µm filter using a liquid to solid (L/S) ratio of approximately 10 in order to remove salts 2) a second portion was subjected to electrodialytical remediation for removal of heavy metals and salts (unpublished

results, process similar to Kirkelund et al., 2010). In the following the three fly ash batches are named 1) fly ash (untreated), 2) fly ash (washed) and 3) fly ash (ED).

The Nalunaq tailings were washed in a 45 µm filter with distilled water using a L/S ratio of approximately 10 in order to remove potential soluble cyanide. Samples of the unwashed and washed Nalunaq tailings were analysed for total cyanide according to DS/ISO 17380:2005.

All materials were dried at 105°C for 48 hours.

2.2.2 Characterisation of samples

The particle size distributions were determined on a Malvern Mastersizer 2000 laser diffractometer. All samples, except the marine clay, were analysed in dry state. Due to agglomeration of clay sized particles, the marine clay sample was instead pre-dispersed in approximately 20 mL 0,005 M $Na_4P_2O_7$ and analysed by wet dispersion in deionized water. For all samples, the Mie theory was applied in the calculations of the particle size distribution using a real refractive index of 1.5 and an imaginary refractive index of 0.1.

Major-element analyses were determined using energy dispersive X-ray fluorescence (ED-XRF) on pressed powder tablets prepared from crushed samples. The loss on ignition (LOI) was based on weight loss between 105°C and 1000°C and was measured after heating at 1000°C for 2hours. The qualitative mineralogy was analysed by X-ray powder diffraction (XRPD) on a X'Pert PRO diffractometer, using Ni-filtered Cu K_α radiation generated at 45 kV and 40 mA.

Leaching tests were conducted on all samples, except the marine clay, according to the principles stated in DS/EN 12457-1 (2002). 40.0 g of material was placed in a bottle and 80.0 mL distilled water was added in order to achieve a liquid to solid ratio of 2. The suspension was agitated for 24 hours. Afterwards, the suspension settled for 15 minutes and was filtrated through a 45 µm filter. The resulting eluates were measured by inductively coupled plasma optical emission spectrometry (ICP-OES) for As, Ba, Cd, Cr, Cu, Mn, Na, Ni, Pb, and Zn.

The total heavy metal concentrations (As, Cd, Cr, Cu, Ni, Pb and Zn) of ashes and tailings were measured by ICP-OES after pre-treatment according to Danish standard DS 259 (2003), where 1.0 g of dry sample and 20.0 mL 7.3 M HNO_3 were heated at 200 kPa (120 °C) for 30 min. The liquid was separated from the solid particles by vacuum through a 0.45 µm filter and diluted to 100 mL.

2.2.3 Production and test of brick-pellets

Torres et al. (2009) demonstrated that initial tests conducted on small scale brick pellets were a useful screening tool for selecting the best compositions of mixtures of clay and waste. In this paper a similar approach was adopted, using the same pellet size, moisture content, dry-pressing load and maximum firing temperature as described by Torres et al. (2009). Brick-pellets were prepared from different mixtures of samples (see Table 1).

Mixture name	Marine clay (%)	Fly ash - untreated (%)	Fly ash - washed (%)	Fly ash - ED (%)	Bottom ash (%)	Nalunaq (%)	Tanbreez fines (%)	Tanbreez mafic (%)
1	100							
2A	80	20						
2B	60	40						
3A	80		20					
3B	60		40					
4A	80			20				
4B	60			40				
5A	80				20			
5B	60				40			
6A	80					20		
6B	60					40		
7A	80						20	
7B	60						40	
8A	80							20
8B	60							40

Table 1. Overview of pellet mixtures with amounts given in wt %.

The Atterberg limits (liquid and plastic limits) were determined, in order to investigate the plastic properties of the mixtures. The liquid limits (w_L) were measured using the Casagrande method (DIN 18.122-1, 1997) and plastic limits (w_P) were determined by the rolling method (DS/CEN ISO/TS 17892-12, 2004).

For the preparation of the pellets, 2 g of the mixture was placed in a small vial and distilled water was added in order to obtain a moisture content of approximately 15%. The vials were sealed and the mixtures were left overnight to absorb the moisture. Pellets were produced by uniaxial compression in a purpose-built pellet mould placed in a manually operated Stenhøj hydraulic press. A maximum load of 14.81 ± 0.04 kN (equivalent to a pressure of 46.5 ± 0.2 MPa) was applied. The initial weights, diameters and heights of the pellets were measured by use of a laboratory scale and a precision micrometer scale gauge. The pellets were dried at 105°C for 24 hours before the weights, diameters and heights were measured again. The pellets were fired at 1000°C for 1 hour in a Vecstar laboratory Furnace, using an average heating and cooling rate of 6.8°/min ± 2.5°/min and 1.9°/min ± 0.5°/min, respectively. After cooling to approximately 200°C, the pellets were removed from the furnace and placed in a desiccator in order to cool to room temperature before weights, diameters and heights were measured again.

A total of seven pellets were produced from each mixture. Three were used for measurements of open porosity, dry density and vacuum water absorption, three were used for leaching tests and one pellet was used for mineralogy.

The open porosity, dry density and vacuum water absorption were determined following the principles of procedure Ti-B-25 (1983, in Danish) by the Danish Technological Institute: The pellets were dried at 105°C, cooled to room temperature in a desiccator and weighed (m_{dry}). The dried pellets were then placed in a desiccator under vacuum for approximately 3 hours. After 3 hours, deionised water at room temperature was led into the desiccator, so that the pellets were completely submerged. Vacuum was maintained for this condition for 1 hour. Hereafter, air was let into the desiccator and the submerged pellets were left at atmospheric pressure overnight. The water saturated pellets were weighed in water (m_{sw}) and in air after wiping excess water of the surface (m_{sa}). The different parameters were calculated as described in the following:

Open porosity $= \frac{m_{sa} - m_{dry}}{m_{sa} - m_{sw}} \cdot 100\%$

Dry density $= \frac{m_{dry} \cdot \rho_w}{m_{sa} - m_{sw}} \cdot 100\%$, where ρ_w is the density of water at room temperature.

Vacuum water absorption $= \frac{m_{sa} - m_{dry}}{m_{dry}} \cdot 100\%$

Leaching tests on crushed pellets, which would represent waste bricks or worst-case scenario leaching, were conducted according to the principles stated in DS/EN 12457-1 (2002). Three pellets from each mixture were crushed and homogenised in order to obtain enough material (approximately 5-6 g) for one leaching test. The crushed pellets were placed in a bottle and distilled water was added in order to achieve a liquid to solid ratio of 2. The suspension was agitated for 24 hours. Afterwards, the suspension settled for 15 minutes and was filtrated through a 45 µm filter. The resulting eluates were measured by ICP-OES for As, Ba, Cd, Cr, Cu, Mn, Na, Ni, Pb, and Zn.

The qualitative mineralogy of the crushed pellets was investigated by XRPD using the same equipment and operating conditions as described in section 2.2.2

4. Results

The particle size distributions of all samples are presented in Figure 1. The fly ash (washed and ED) and the Tanbreez fines had similar particle size distributions as the marine clay. In comparison, the fly ash (untreated) contained a larger volume % of fine particles; whereas the bottom ash, Nalunaq tailings and Tanbreez mafic were dominated by coarser particles.

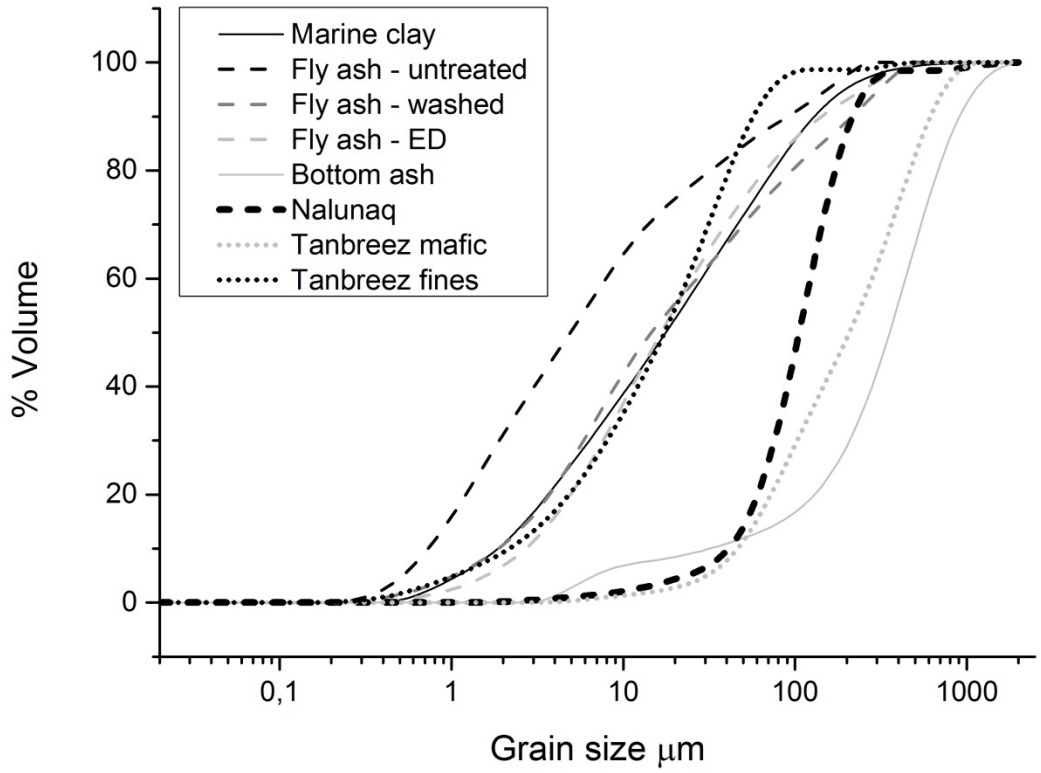

Figure 1. The particle size distributions of the investigated raw samples.

	Marine clay	Fly ash - untreated	Fly ash - washed	Fly ash - ED	Bottom ash	Nalunaq	Tanbreez fines	Tanbreez mafic
SiO_2	61.12	4.53	11.87	9.29	35.97	50.71	50.56	46.76
TiO_2	0.59	0.85	1.57	1.38	1.47	0.90	0.48	2.19
$Al2O_3$	14.52	2.96	6.92	5.27	7.35	13.03	16.69	7.43
Fe_2O_3T	6.48	1.19	2.25	1.63	5.02	11.71	8.68	29.69
MnO	0.08	0.04	0.08	0.07	0.16	0.14	0.46	0.56
MgO	3.52	0.58	2.08	1.89	1.66	5.36	0.00	0.54
CaO	2.36	23.20	36.62	26.33	38.97	15.08	6.95	4.13
Na_2O	3.31	14.90	4.94	6.41	2.03	0.73	11.31	7.37
K_2O	2.81	8.21	1.64	3.74	1.06	0.78	2.53	1.32
P_2O_5	0.12	0.55	1.92	1.90	1.73	0.00	0.00	0.00
LOI	5.09	43.01	30.10	42.11	4.58	1.58	2.33	0.01

Table 2. The major element distribution and loss on ignition (LOI) given as wt % of the investigated raw samples.

The major element composition and loss on ignition data are presented in Table 2. The marine clay, Nalunaq and Tanbreez tailings were dominated by SiO_2. Al_2O_3 was also a major component, however for Nalunaq and Tanbreez mafic, CaO and Fe_2O_3T (i.e. total Fe_2O_3), respectively, were more dominant than Al_2O_3. The fly ashes and bottom ash were dominated by CaO, although the bottom ash also contained a large fraction of SiO_2. The fly ashes had large LOI values, ranging from

30-43 weight %. The amounts of the fluxing oxides, Na_2O, K_2O, which lower the melting temperature during firing (Worrall, 1975) ranged from 1.51-23.11 wt % with the highest values found for the fly ash (untreated) and Tanbreez fines.

The total cyanide measured for the washed and unwashed Nalunaq samples were very similar with values of 26 mg/kg and 18 mg/kg, respectively. In comparison, a level of 50 mg/l weak acid dissociable (WAD) cyanide (which constitutes parts of the total cyanide concentration) has been used as regulatory guideline for tailings slurries in open disposals in the United States and Australia. This level is acceptable for animals, which come into contact with the tailings (Lottermoser, 2007). The initial washing did not contribute to any reduction of cyanide, and could therefore have been emitted. The results of the XRPD analyses of the raw samples are presented in Table 3 and the diffractograms of the raw samples and pellets are compared in Figure 2.

	Mineral phases
Marine clay	Amphibole, chlorite, feldspar, mica (biotite), pyroxene, quartz
Fly ash - untreated	Anhydrite, feldspar, calcite, halite, sylvite
Fly ash - washed	Anhydrite, calcite, feldspar, halite, hydrate?, quartz
Fly ash - ED	Anhydrite, calcite, feldspar, halite, sylvite, quartz
Bottom ash	Calcite, CaO, CaOH, larnite/belite, feldspar, quartz
Nalunaq	Ca-Amphibole, Ca-feldspar, pyroxene, quartz, chlorite, biotite
Tanbreez fines	Aenigmatite, eudialyte, Na-Amphibole, Na/K-feldspar, Na-pyroxene, natrolite, sodalite
Tanbreez mafic	Aenigmatite, eudialyte, Na-Amphibole, Na/K-feldspar, Na-pyroxene, sodalite

Table 3. Identified mineral phases from XRPD. Mineral phases are listed alphabetically

As expected the marine clay had the dominant influence on the mineralogical composition of the pellets, due to its larger weight fraction, and all pellets contained quartz and feldspar. Chlorite and mica peaks were not observed in the pellet diffractograms, indicating that these phases were decomposed at temperatures below 1000°C. The same was generally also observed for amphibole, although for the Nalunaq pellets (6A and 6B) amphibole was still present after firing.

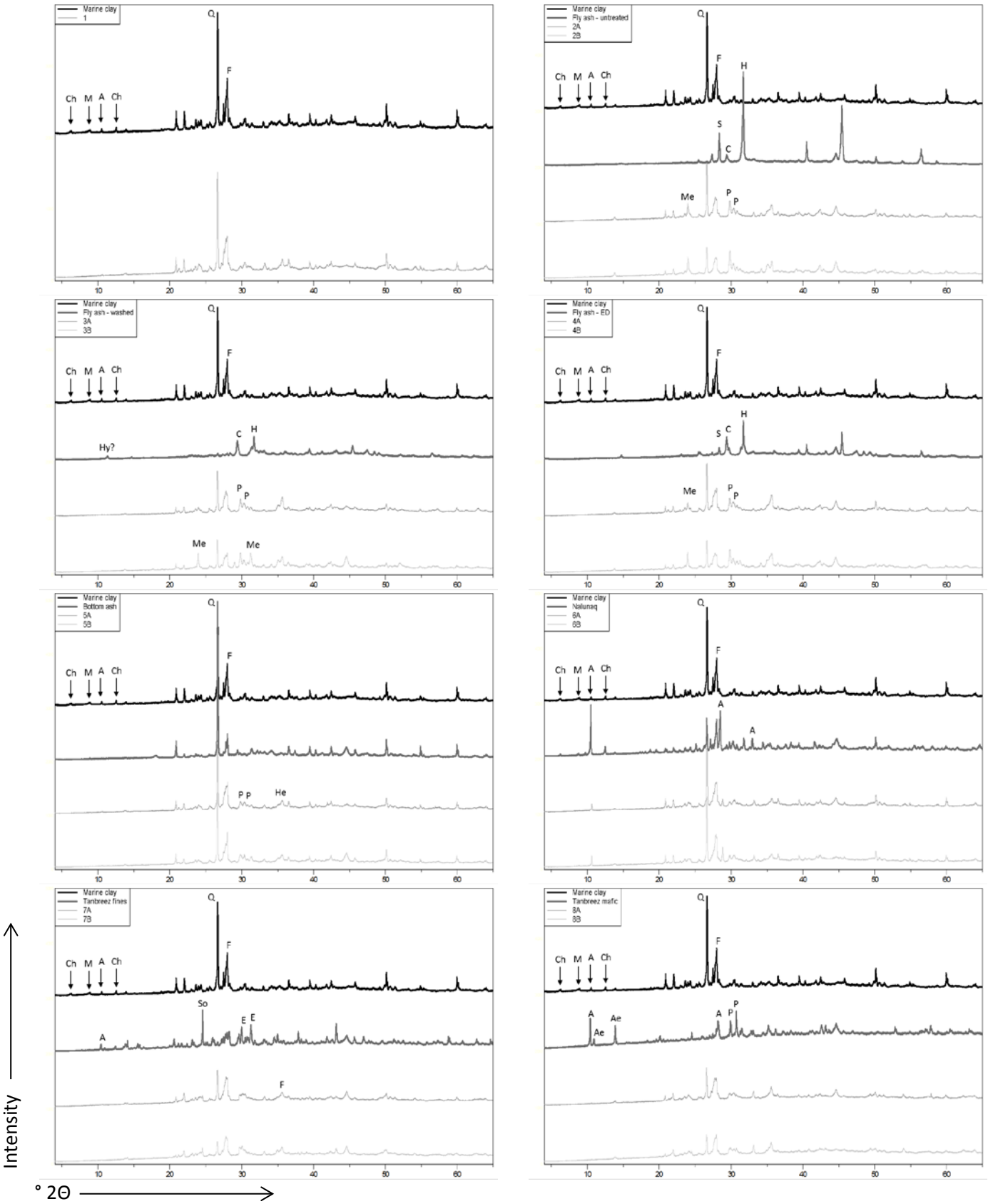

Figure 2. Comparison of XRPD diffractograms of related raw samples and pellets. A=amphibole, Ae=aenigmatite, C=calcite, Ch=chlorite, F=feldspar, H=halite, He=hematite, Hy=hydrate, M=mica, Me=melilite, P=pyroxene, S=sylvite, So=sodalite

All pellets showed peak shifts due to high temperature transitions of feldspars, e.g. microcline to sanidine. For the fly ash pellets, melilite and pyroxene (diopside) peaks were seen to appear after firing, whereas halite (NaCl), sylvite (KCl) and calcite ($CaCO_3$) peaks were not observed after firing. The Tanbreez tailings were similar in their qualitative mineralogy and contained several Na-rich mineral phases, such as aegirine (Na-pyroxene), Arfvedsonite (Na-amphibole), aenigmatite and sodalite. The diffractograms of the 7B and 8B pellets (40% fines and mafic) showed markedly reduced quartz peaks compared with the other diffractograms, possibly indicating enhanced decomposition of quartz.

The total metal concentrations and leaching results from the investigated samples and pellets are shown in Table 4. There are currently no Greenlandic regulations or guidelines for incorporating waste in construction materials or for using it for construction purposes. The Danish (or European) regulations, which are often adapted in Greenland when local guidelines are lacking, only include regulations for reuse of waste for construction purposes, i.e. in roads, dams, foundations etc. (the Danish Ministry of the Environment, 2010) and does not apply to hazardous materials (classified according to the Danish Ministry of the Environment (2012) and the European parliament and the council of the European Union (2008)). Although, the investigated waste materials and crushed pellets might classify as hazardous, it is still informative to compare with the Danish regulation values as a first approximation. The categories 1, 2 and 3 of the Danish regulation (Danish Ministry of the Environment, 2010) are controlled by the total and leached concentrations and the values shown in Table 4 represent maximum allowed values in each category. A material as a whole is categorised according to the highest category obtained for the individual elements, ions or compounds.

Waste materials

	Fly ash untreated	Fly ash washed	Fly ash ED	Bottom ash	Nalunaq	Tanbreez fines	Tanbreez mafic	Category 1	Category 2	Category 3
Total concentration (mg/kg)										
As	21 ± 1	228 ± 4	942 ± 73	10 ± 2	156 ± 10	<2	<2	0-20	> 20	> 20
Cd	93 ± 3	244 ± 3	827 ± 25	5 ± 4	<2	<2	<2	0-0.5	> 0.5	> 0.5
Cr	70 ± 2	196 ± 2	195 ± 5	64 ± 2	36 ± 1	<2	<2	0-500	> 500	> 500
Cu	450 ± 10	2700 ± 19	2160 ± 15	792 ± 542	128 ± 3	<2	<2	0-500	> 500	> 500
Ni	47 ± 4	55 ± 1	61 ± 2	40 ± 6	63 ± 1	<2	<2	0-30	> 30	> 30
Pb	890 ± 34	10200 ± 68	6750 ± 81	449 ± 187	11 ± 2	13 ± 0	11 ± 3	0-40	> 40	> 40
Zn	6870 ± 416	53100 ± 1021	59700 ± 713	2410 ± 470	52 ± 5	33 ± 0	36 ± 1	0-500	> 500	> 500
Leaching (µg/l)										
As	1240 ± 122	<20	488 ± 40	22 ± 2	38 ± 9	<20	<20	0 - 8	0 - 8	8-50
Ba	2670 ± 33	257 ± 19	596 ± 16	13600 ± 406	<20	<20	<20	0-300	0-300	300-4000
Cd	1460 ± 106	<20	580 ± 51	<20	<20	<20	<20	0-2	0-2	2-40
Cr	11500 ± 688	3060 ± 345	1240 ± 46	265 ± 9	<20	<20	<20	0-10	0-10	10-500
Cu	74 ± 5	<20	80 ± 7	75 ± 18	<20	33 ± 3	20 ± 7	0-45	0-45	45-2000
Mn	<20	<20	<20	<20	<20	<20	73 ± 29	0-150	0-150	150-1000
Na ($\cdot 10^5$)	418 ± 4	95 ± 3	222 ± 4	9.3 ± 0.2	0.03 ± 0.001	0.8 ± 0.08	0.3 ± 0.03	0-1	0-1	1-15
Ni	<20	<20	<20	<20	<20	<20	<20	0-10	0-10	10-70
Pb	455 ± 64	30 ± 15	421 ± 75	2460 ± 507	<20	<20	69 ± 40	0-10	0-10	10-100
Zn	1070 ± 190	126 ± 11	257 ± 76	1710 ± 174	<20	47 ± 3	252 ± 102	0-100	0-100	100-1500

Pellets

	2A	2B	3A	3B	4A	4B	5A	5B	6A	6B	7A	7B	8A	8B
Leaching (µg/l)														
As	307	464	286	163	1250	1390	119	56	534	582	131	145	<20	65
Ba	26	526	284	349	103	633	54	342	21	42	75	68	104	75
Cd	<20	<20	<20	<20	<20	24	<20	<20	<20	<20	<20	<20	<20	<20
Cr	4170	21800	5700	8900	4270	13400	3490	13700	291	357	1200	970	979	804
Cu	40	47	28	37	53	37	60	25	32	22	41	113	74	76
Mn	<20	31	<20	<20	<20	25	<20	<20	<20	<20	109	154	109	125
Na ($\cdot 10^5$)	6.0	4.6	2.8	6.1	4.6	32.0	1.7	1.9	0.9	0.8	1.6	1.7	1.5	1.2
Ni	<20	21	<20	<20	20	<20	<20	<20	<20	<20	<20	39	<20	24
Pb	630	<20	<20	96	<20	<20	<20	<20	<20	<20	121	45	89	49
Zn	322	117	51	68	412	137	775	58	68	48	80	733	263	501

Table 4. Total metal concentrations of the raw samples and leaching properties (L/S=2) of the raw samples and pellets. Values are compared to Danish regulatory values (category 1-3) for construction purposes (Danish Ministry of the Environment, 2010).

When comparing the leaching tests and total concentrations measured for the investigated waste samples in this study with the regulation, it was apparent that all the fly ashes and the bottom ash classified outside category due to high concentrations of e.g. As, Cd and Cr, and were not suitable for use according to the regulation. The Nalunaq and Tanbreez mafic tailings both classified as category 3, due to high leaching values of As for Nalunaq and Pb and Zn for Tanbreez mafic. Most of the values for Tanbreez fines comply with category 1, however due to standard limitations it was not possible to determine the exact total concentration of Cd and leaching values for As, Cd, Cr, Ni and Pb and it was therefore not possible to determine, which of the three categories it belonged to. Non-hazardous waste bricks are also covered by the Danish regulation for reuse of waste for construction purposes. However, according to the leaching tests all crushed pellets, which represent waste bricks, classified outside category. Although the pellets only contained 20% and 40% of the waste materials, leaching values for some species were observed to increase in comparison to the raw samples. Particularly, As and Cr became more available to leaching after firing.

The Atterberg limits (plastic and liquid limits) and the plasticity index are given in Table 5.

Mixture name	w_P(%)	w_L (%)	PI
1	24.2	29.8	5.6
2A	24.2	30.0	5.8
2B	25.2	29.6	4.4
3A	31.1	36.3	5.2
3B	37.9	45.4	7.5
4A	34.0	43.6	9.6
4B	45.7	53.1	7.4
5A	25.3	29.1	3.8
5B	25.6	28.7	3.1
6A	20.2	24.2	4.0
6B	19.0	22.3	3.3
7A	23.6	29.6	6.0
7B	22.0	27.7	5.7
8A	19.7	25.6	5.9
8B	15.0	20.1	5.1

Table 5. The plastic limit (w_P), liquid limit (w_L), and plasticity index (PI, calculated as w_L - w_P) of the pellet mixtures (before firing).

The plastic limit (w_p) of a mixture represent the lower boundary of plastic behaviour, i.e. the water content above which the mixture can be moulded and retain its shape. As the liquid limit (w_L) is exceeded the mixtures will no longer be coherent and lose their plasticity. The plasticity index (PI) is included to evaluate the span between the plastic and liquid limit. All mixtures had low plasticity indices, which indicated that their 'moulding range' was very limited. The fly ash and bottom ash mixtures generally had both plastic and liquid limits in the same range or above that of the pure clay (mixture 1), whereas the tailings generally had lower limits compared to the pure clay.

The initial water content of the pellets before firing and the shrinkage, appearance, water absorption, open porosity and dry density after firing are shown in Table 6. Some pellets had negative firing shrinkages, i.e. they expanded slightly during firing without their overall shape and coherency being affected. The open porosity increased with addition of fly ash and decreased with addition of bottom ash and mine tailings in comparison to the natural clay. The same trend was also observed for the water absorption.

Mixture name	Initial water content	Drying Shrinkage	Firing Shrinkage	Appearance of fired pellets	Vacuum water absorption (%)	Open porosity (%)	Dry density (kg/m^3)
1	15.8 ± 1.1	0.3 ± 0.1	0.5 ± 0.1	Red	23.3 ± 0.2	39.5 ± 0.3	1691 ± 3
2A	14.9 ± 1.5	0.0 ± 0.0	0.1 ± 0.3	Red/yellow, spotted appearance	26.7 ± 1.4	42.1 ± 1.3	1578 ± 34
2B	13.4 ± 1.0	0.2 ± 0.1	0.7 ± 0.2	Yellow, few red spots	32.2 ± 1.5	47.2 ± 1.4	1461 ± 27
3A	15.2 ± 1.1	0.3 ± 0.1	-0.3 ± 0.1	Red/yellow, spotted appearance	29.6 ± 0.4	45.8 ± 0.4	1544 ± 8
3B	17.2 ± 2.2	0.4 ± 0.1	-0.4 ± 0.1	Yellow, few red spots	34.4 ± 0.7	49.9 ± 0.4	1446 ± 18
4A	15.6 ± 1.0	0.3 ± 0.1	-0.2 ± 0.1	Red/yellow, spotted appearance	32.9 ± 1.1	47.9 ± 0.9	1453 ± 20
4B	16.5 ± 1.6	0.2 ± 0.1	0.1 ± 0.1	Yellow/red, spotted appearance	39.2 ± 0.3	51.5 ± 1.2	1313 ± 22
5A	14.7 ± 1.2	0.3 ± 0.2	-0.1 ± 0.1	Light red	23.9 ± 0.8	39.9 ± 1.0	1669 ± 21
5B	14.8 ± 0.4	0.3 ± 0.1	-0.2 ± 0.0	Yellow/light red	22.5 ± 0.6	38.4 ± 0.9	1708 ± 8
6A	15.0 ± 0.4	0.3 ± 0.1	0.1 ± 0.0	Red	19.6 ± 0.4	35.5 ± 0.8	1808 ± 5
6B	13.1 ± 1.2	0.1 ± 0.2	-0.6 ± 0.1	Red	18.0 ± 0.6	33.8 ± 0.8	1868 ± 20
7A	15.4 ± 0.5	0.2 ± 0.2	0.9 ± 0.1	Red	21.5 ± 0.9	37.2 ± 1.5	1728 ± 20
7B	15.8 ± 0.5	0.1 ± 0.1	2.0 ± 0.2	Dark red	19.3 ± 0.2	34.6 ± 0.3	1789 ± 21
8A	13.3 ± 0.7	0.6 ± 0.1	0.0 ± 0.1	Red with black crystals	18.7 ± 1.1	34.0 ± 1.6	1819 ± 26
8B	13.6 ± 0.9	0.4 ± 0.2	0.5 ± 0.4	Dark red with black crystals	13.9 ± 0.2	27.9 ± 0.2	2007 ± 10

Table 6. The initial water content (before drying and firing), shrinkage, appearance, vacuum water absorption, open porosity and dry density of the pellets.

5. Discussion

5.1 Mixing and choice forming process

Based on a spotted appearance of the fired fly ash pellets it was clear that the mixtures of clay and fly ashes had not been homogenised fully during mixing. For larger scale testing or production, the application of mechanical grinding or milling in the mixing process could potentially solve this problem.

A previous study by Belmonte et al. (2014, b) reported a plastic and liquid limit of 20.5% and 30.2%, respectively, for the same marine clay as used in this study. The liquid limits were nearly identical (29.8 % in this study) but the plastic limit in this study was 24.2 %, indicating that the initial treatment by drying at 105°C had increased the plastic limit. This is contradictory to the decrease in plastic limit with increasing temperature, which has been reported for other types of clay (Abu-Zreig et al., 2001; Basma et al., 1994). Agglomeration of clay particles during drying followed by incomplete disaggregation could be a likely explanation, although this would have to be confirmed. The low plasticity index (< 10) found for all tested raw samples indicate that their plastic range is very limited, which can present a problem in the forming process. In the case of the dry pressing method (which was used in this study) the plasticity is not important, but plays an important role in other forming methods, such as the extrusion or soft-mud methods (Brick Industry Association, 2006). The high plastic limits of the fly ashes compared to the other raw samples indicate that more water would be needed in order to obtain a mouldable mass. More water would, however, also increase the shrinkage and thereby the risk of developing cracks during firing.

5.2 Physical properties and mineral chemistry of the pellets

The small negative firing shrinkage (expansion) observed for pellets 3A (20% fly ash (washed)), 3B (40% fly ash (washed)), 4A (20% fly ash (ED)), 5A (20% bottom ash)and 5B (40% bottom ash) is

most likely explained by liberation of gas, e.g. from decomposition of sulphate (from anhydrite) and carbonate (from calcite) at temperatures at or above the sintering temperature, which can cause bloating (Riley, 1951). The expansion is not observed for mixture 4B (40% fly ash ED), however, the above mentioned heterogeneities could account for this. Furthermore, the measurement precision was limited due to the small size of the pellets. The fly ash (untreated) likely contains less calcite compared to the other fly ashes and the bottom ash, based on its lower content of CaO. This difference could also explain why the fly ash (untreated) pellets do not show evidence of expansion. The Nalunaq pellets, 6B, had negative firing shrinkages, whereas pellets 6A, had positive firing shrinkages. Apparently, expansion effects therefore occur for compositions containing more than 20 % of the Nalunaq tailings. The presence of sulphides, which are associated with the Nalunaq ore (Schlatter and Olsen, 2011), could give rise to bloating effects as SO_2 is liberated during firing (Riley, 1951). Compositons 8A and 8B (20% and 40% Tanbreez mafic, respectively) have some of the lowest observed open porosities and highest densities of all investigated pellet types, indicating a high degree of sintering. The high degree of sintering is consistent with the high content of Na compounds, which can act as fluxing materials and aid vitrification at elevated temperatures (Bloodworth et al., 2007). Furthermore the observed reduction of the quartz peaks is also consistent with enhanced melt formation, i.e. vitrification. Interestingly the firing shrinkages of compositions 8A and 8B seem to be lower than that of the clay, although the drying shrinkages are slightly higher. Dry densities of clay bricks, which are used in the building industry have been reported to be in the range of 1610-2120 kg/m^3 (Dondi et al., 2004; Dondi et al., 2000) and open porosities in the range of 18.8-39 vol % (Dondi et al., 2004). In comparison to these values the fly ash mixtures generally have lower densities and higher porosities, whereas the clay, bottom ash and tailings mixtures are within the range. Because of the high open porosity and water absorption of the fly ash pellets, it is likely that these will be less durable compared to the other pellet types, due to e.g. spalling effects created by saline water

mobility or freezing and thawing. The fly ash could possibly be utilised for lightweight bricks, which should not be in direct contact with water or exposed to outdoor climate.

5.3 Leaching and total concentrations of heavy elements

The increased leaching of heavy metals from the crushed pellets compared to the samples can be problematic. Other authors (Rambaldi et al., 2010; Zhang et al., 2007; Zhang et al., 2011) found that leaching from uncrushed waste-containing ceramics was reduced after sintering, as mobile heavy metals were stabilised in the amorphous phase. If that is the case, this study would either imply that the heavy metals are easily mobilised by crushing or that optimal glass formation for stabilising heavy metals has not been achieved at the tested firing temperature. Furthermore, for the crushed pellets investigated in this study, it would appear that additional heavy metals have been mobilised during firing. An explanation for this could, for example, be the decomposition of sulphides, which (although not detected by XRD) could be present in small quantities and which often contain heavy metals (such as e.g. arsenopyrite, FeAsS). Even though most leaching values appear to be lower for the pellets than the samples, it is important to consider the lower content of waste materials (e.g. 20% and 40%) in the pellets, which for some elements give a false impression of lowered mobility after firing.

5.4 Remediation and upgrading

The remediation techniques applied to the fly ash, e.g. washing and electrodialytical treatment, generally increased the total heavy metal concentrations of the solids, due to removal of soluble salts (as evidenced by Na, Table 4). Heavy metal leaching was reduced for both the remediated ashes compared to the fly ash (untreated). The washing technique led to the highest reduction in leaching concentrations of all measured heavy metals, except Cr, which was reduced more using the electrodialytical treatment. Neither of the fly ashes did, however, meet the requirements of

the Danish regulations for reuse of waste for construction purposes (the Danish Ministry of the Environment, 2010). Depending on future regulatory values, the possibilities for remediating or upgrading all the studied waste materials should therefore be further investigated. A study by Kirkelund et al. (2012) on the MSWI bottom ash from Sisimiut for use as an aggregate replacement in road construction, found that leaching of heavy metals, such as Cu and Cr were higher in the finest fractions. They concluded that the removal of these fractions by sieving would give a material which was less problematic and could be used for geotechnical purposes. A similar investigation should be attempted for the tailings and the influence of gravity separation could also be tested in order to remove e.g. sulphides. For all the investigated materials, the vitrification treatment (e.g Colombo et al., 2003), where materials are fused at high temperatures in order to fix heavy metals in a glass matrix, could also be applied before incorporation.

5. Conclusions

The open porosities and densities of the tailings and bottom ash pellets were acceptable for brick production. The fly ash pellets, regardless of pre-treatment, generally had problematic properties, such as high open porosity, high water absorption and negative firing shrinkage compared to the other waste materials. These properties indicate that bricks incorporating the fly ashes could be less durable. Leaching values of all the tested pellets generally revealed a higher mobility of As and Cr after firing, which was likely caused by decomposition of As and Cr containing phases, e.g. sulphides, during firing. For the fly ash, the two applied remediation techniques, washing and electrodialytical treatment, reduced the leaching from the samples. None of fly ashes, however, met the required levels of heavy metals according to the Danish regulations for reuse of waste for construction purposes. This study concludes that initial or additional remediation of all samples should be investigated further.

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Appendix 6

Description: Unpublished paper

Title: Optimisation of firing temperature for marine clay from Ilulissat

Optimisation of firing temperature for marine clay from Ilulissat

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Abstract

The effect of the firing temperature on the properties of sintered brick pellets produced from marine clay from Ilulissat, West Greenland was investigated in order to optimise the firing temperature. The sintering behaviour was evaluated on basis of the variation in firing shrinkage, open porosity, bulk density and vacuum water absorption, while the mineralogical phase changes were investigated by XRD. The investigation was based on firing temperatures of 950°C, 1000°C and 1050°C. When the firing temperature was increased, the firing shrinkage and bulk density increased and the open porosity and water absorption decreased, indicating increased sintering. However, the changes were larger in the range from 1000-1050°C compared to 950-1000°C. The mineralogical investigations showed that chlorite, mica and amphibole were decomposed at temperatures below 1050°C. The study concluded that the optimum firing temperature was above 1000°C.

Keywords: marine clay, ceramics, optimisation of firing temperature

1. Introduction

The optimisation process is an important part of producing clay-based ceramics, such as bricks and tiles, in order to determine the optimal firing temperature. During the firing process, the raw materials sinters, such that particles are joined and the mechanical strength increases (Reed,

1995). Depending on the physical and chemical properties of the raw materials, i.e. grain size distribution, major element chemistry etc., the firing process greatly influences parameters such porosity, density, water absorption and strength (Brick Industry Association, 2006; Cultrone et al., 2004; Karaman et al., 2006). According to the Brick Industry Association (2006), sintering generally takes place in the temperature range of 871-1316°C. Temperatures in the range of 900-1100°C, are, however, standardly applied for initial investigations on bricks (Bloodworth et al., 2007; Dondi et al., 1997a,b; Zhang, 2013).

Belmonte et al. (2014) demonstrated that marine clay from Ilulissat, West Greenland was highly suited for production of bricks. However, the study concluded that the applied firing temperatures of 1045-1080°C, lead to a high degree of sintering and the possibility for producing good quality bricks at lowered temperatures should therefore be investigated. In this paper, small test pellets were, therefore, prepared from the same Ilulissat clay as used by Belmonte et al. (2014) and were fired at temperatures of 950°C, 1000°C and 1050°C in order to determine the influence of the firing temperature on properties such as open porosity, bulk density and water absorption.

2. Material and Methods

Approximately 150 kg of marine clay from Ilulissat, West Greenland was sampled from a depth interval of 20 cm – 100 cm below surface. The material was roughly homogenised using a large shovel and distributed into 10 L plastic buckets, which were sealed and stored at room temperature until use.

The < 1000 µm fraction of the clay was obtained by wet sieving and subsequently dried at 105°C and lightly disaggregated using a mortar and pestle. 2 g of the dried and disaggregated material was placed in a small vial and distilled water was added in order to obtain a moisture content of 15 ± 1%. The vials were sealed and the materials were left overnight to absorb the moisture. Small

bricks-pellets were produced by uniaxial compression in a purpose-built pellet mould placed in a manually operated Stenhøj A/S hydraulic press. A maximum load of 14.80 ± 0.04 kN, equivalent to a pressure of 46.4 ± 0.2 MPa, was applied. The production of pellets or powder discs using similar loads in order to conduct preliminary investigations of ceramic properties had previously been employed succesfully by Romero et al. (2008) and Torres et al. (2009). The initial diameters and heights of the pellets were measured by a precision micrometer scale gauge. The pellets were dried at 105°C for 24 hours before the diameters and heights were measured again. Pellets were fired at 950°C, 1000°C and 1050°C using a soaking time of 1 hour in a Vecstar laboratory Furnace. The firing curves can be seen in Figure 1. After cooling to approximately 200°C, the pellets were removed from the furnace and placed in a desiccator in order to cool to room temperature before diameters and heights were measured again.

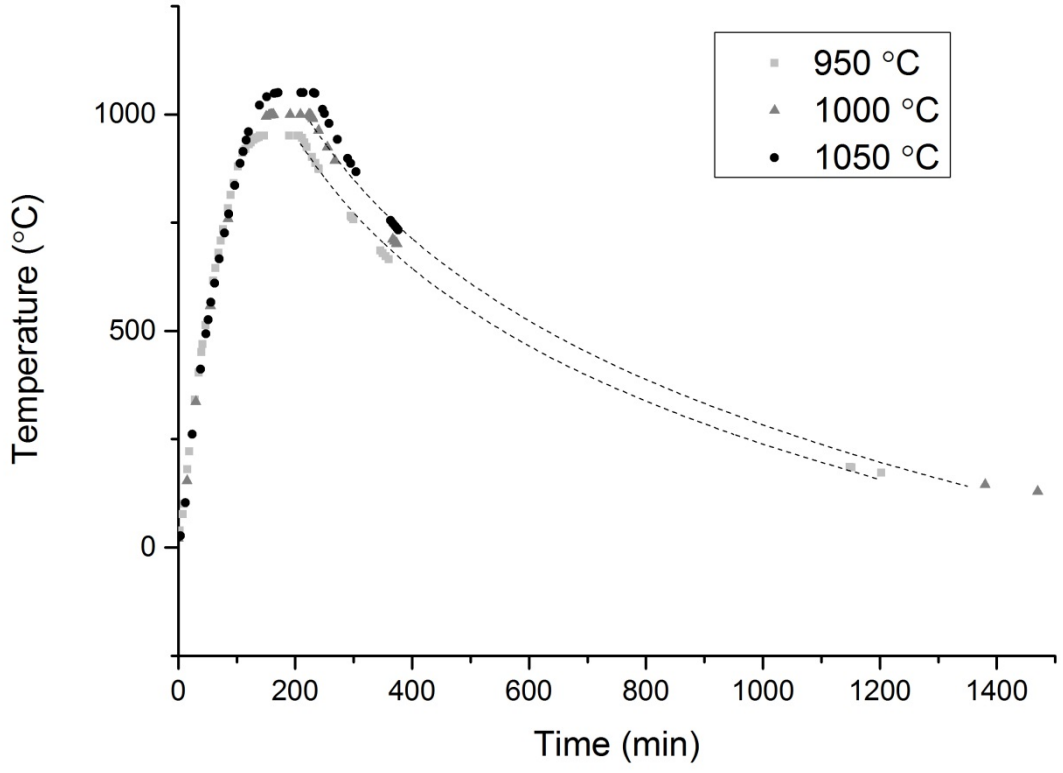

Figure 1. Firing curves for the marine clay pellets. The dotted lines represent the estimated cooling curves, which were calculated using logarithmic functions.

Four pellets were produced for each sample: three were used for the determination of open porosity, bulk (apparent) density and vacuum water absorption and one for the mineralogical investigations. The porosity, density and vacuum water absorption were determined according to procedure TI-B-25 (1983) by the Danish Technological Institute. The method is briefly described in the following. The pellets were dried at 105°C, cooled to room temperature in a desiccator and weighed (m_{dry}). The dried pellets were then placed in a desiccator under vacuum for approximately 3 hours. After 3 hours, de-ionised water at room temperature was led into the desiccator, so that the pellets were completely submerged. Vacuum was maintained for this condition for 1 hour. Hereafter, air was let into the desiccator and the submerged pellets were left at atmospheric pressure overnight. The water saturated pellets were weighed in water (m_{sw}) and in air after wiping excess water of the pellet surfaces (m_{sa}). The different parameters were calculated as described in the following:

The open porosity $= \frac{m_{sa} - m_{dry}}{m_{sa} - m_{sw}} \cdot 100\%$

Apparent (bulk) density $= \frac{m_{dry} \cdot \rho_w}{m_{dry} - m_{sw}} \cdot 100\%$, where ρ_w is the density of water at room temperature.

Vacuum water absorption $= \frac{m_{sa} - m_{dry}}{m_{dry}} \cdot 100\%$

The bulk mineralogy of the marine clay and fired pellets were analysed by X-ray powder diffraction (XRPD) on a X'Pert PRO diffractometer, using Ni-filtered Cu Kα radiation generated at 45 kV and 40 mA.

3. Results and discussion

The results of the investigations on shrinkage properties, open porosity, vacuum water absorption and bulk density as a function of firing temperature are shown in Table 1 and Figure 2.

Firing Temperature (°C)	Drying shrinkage (%)	Firing shrinkage (%)	Total shrinkage (%)	Open porosity (%)	Bulk density (kg/m^3)	Vacuum water absorption (%)
950	0.5 ± 0.2	0.05 ± 0.04	0.6 ± 0.1	39.7 ± 0.7	1670 ± 18	23.6 ± 0.4
1000	0.3 ± 0.08	0.5 ± 0.2	0.8 ± 0.1	39.5 ± 0.3	1690 ± 3	23.3 ± 0.2
1050	0.4 ± 0.1	2.0 ± 0.7	2.4 ± 0.7	32.6 ± 1.0	1830 ± 18	17.8 ± 0.6

Table 1. Shrinkage properties, open porosity, apparent (bulk) density and vacuum water absorption of the pellets in respect to the different firing temperatures.

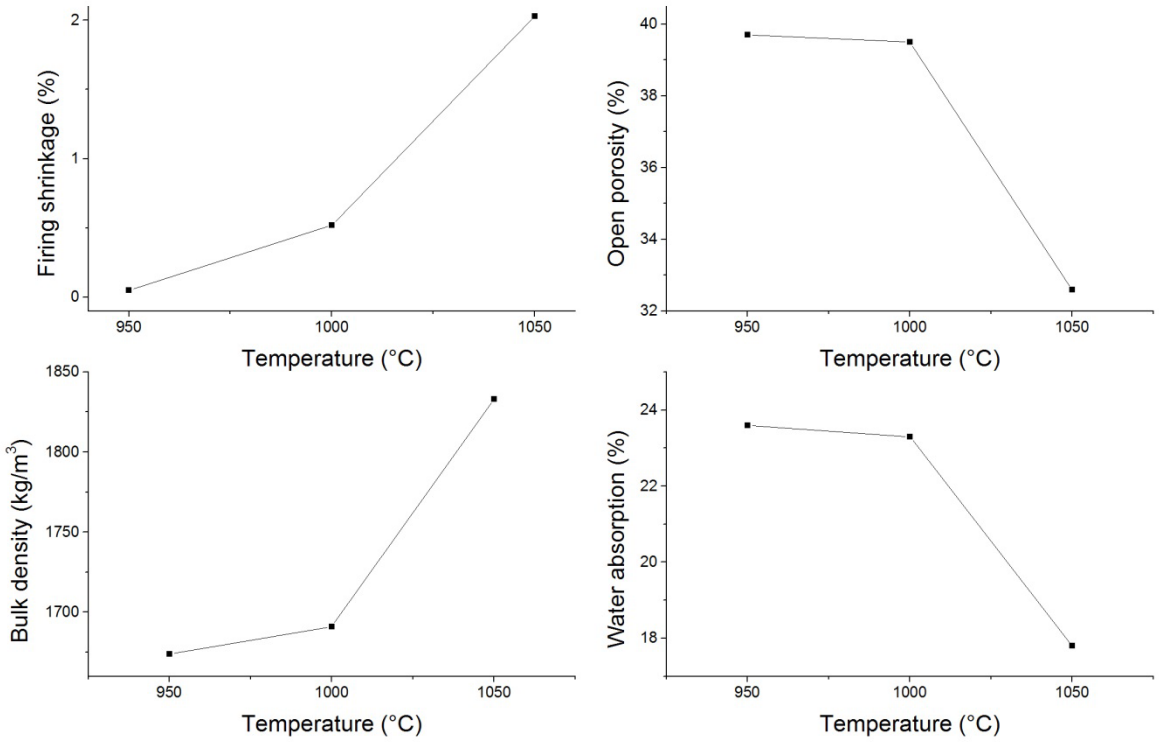

Figure 2. The firing shrinkage, open porosity, bulk density and water absorption of the pellets plotted in respect to the different firing temperatures.

According to the Brick Industry Association (2006) the size shrinkage due to drying and firing usually fall within the ranges of 2-4 % and 2.5-4 %, respectively. In this study, the drying shrinkages are below 1 % and should be roughly the same for all pellets; however, the small variations between the pellets most likely reflect small inhomogeneities in water content, mineralogy and particle size distribution in the individual pellets. In larger scale samples, e.g. normal sized bricks, these variations would be averaged out. Upon firing, the shrinkage increases with temperature indicating a higher degree of sintering for the pellets fired at 1050°C compared to the other

pellets. Furthermore, it is evident that the temperature range of 1000-1050°C involves a larger increase of shrinkage than the range from 950-1000°C (see Figure 2). For the pellets fired at 950°C the firing shrinkage is very low, indicating that no significant sintering takes place below this temperature. Belmonte et al. (2014) reported total shrinkages (i.e. the sum of the drying and firing shrinkages) of 13.8 ± 0.5 % for their marine clay bricks fired at 1045-1055°C. Although the same raw material was used in this study, the total shrinkage is only 2.4 ± 0.7 % for the pellets fired at 1050°C. However, Belmonte et al. (2014) employed the soft-mud forming method, which requires more water and therefore gives larger shrinkage values than the dry-pressing method used in this study (Brick Industry Association, 2006).

Assessing the porosity of bricks is important, as it markedly affects the mechanical properties. Lu et al. (1999) demonstrated, that when the porosity of bricks increases the strength decreases. Open porosities of bricks, which are used in the building industry, have been reported in the range of 18.8-39 vol % (Dondi et al., 2004). The open porosities of the pellets fired at 950-1000°C (39.7 and 39.5 vol %, respectively) are slightly above this range, whereas pellets fired at 1050°C fall within the range (32.6 vol %). As expected the open porosity is seen to decrease with increasing temperature (Cultrone et al., 2004). Furthermore, the temperature range of 1000-1050°C involves a larger decrease of open porosity than the range from 950-1000°C, where no apparent difference is evident. Belmonte et al. (2014) reported an open porosity of 4.9 vol % for their marine clay bricks fired at 1045-1055°C. In the study by Belmonte et al. (2014) the firing programme, however, lasted for > 5 days compared to approximately 1 day in this study, which might explain the difference. Clews (1969) stated that rapid firing could give rise to entrapment of gases (e.g. from dissociation of hydrated minerals (mica) or decomposition of carbonates) or even bloating, which in turn led to a higher porosity of a fired mass. This phenomenon would also reduce the firing shrinkage and provide another explanation for the low total shrinkage observed in this study. It

should, however, be noted that the overall shape of the pellets remained uniform and did not show any signs of bloating.

The vacuum water absorption is directly related to the open porosity and both properties show the same trend, i.e. a larger decrease from 1000-1050°C but no significant decrease from 950-1000°C.

The bulk densities of all pellets are comparable to bulk densities of industrial bricks, which usually are in the range of 1510-2380 kg/m^3 (Dondi et al., 2004; Dondi et al., 2000; Hall et al., 1992). The bulk density is seen to increase with rising temperature, and again the increase is more pronounced in the temperature range from 1000°-1050°C. In general, the data on open porosity, vacuum water absorption and bulk density is consistent with a larger degree of sintering in the 1000°-1050°C range. Belmonte et al. (2014) reported a density of 2350 kg/m^3 for their marine clay bricks fired at 1045-1055°C. Again the differences in the firing programmes might explain the difference from the lower density at 1050°C (1830 ± 18 kg/m^3) found in this study.

The X-ray diffractograms of the marine clay and the pellets at the three firing temperatures are displayed in Figure 3. From the diffractograms, it is evident that the chlorite peaks, which were observed for the marine clay, have disappeared at 950°C. Furthermore the mica and amphibole peaks appear to be diminished at this temperature. The mica peaks disappear between 950-1000°C and the amphibole peaks disappear between 1000-1050°C. These disappearances are attributed to the decomposition of the respective phases at or below the mentioned temperatures. As mica and amphibole decompose, water is liberated and melt formation (vitrification) is enhanced (Cultrone et al., 2004; Gualtieri, 2000). This is consistent with the observed higher shrinkage, higher density, lower porosity and lower water absorption at 1050°C.

For all fired compositions, peak shifts towards high temperature phases are observed for the feldspars. This observation is consistent with observations by Belmonte et al. (2014).

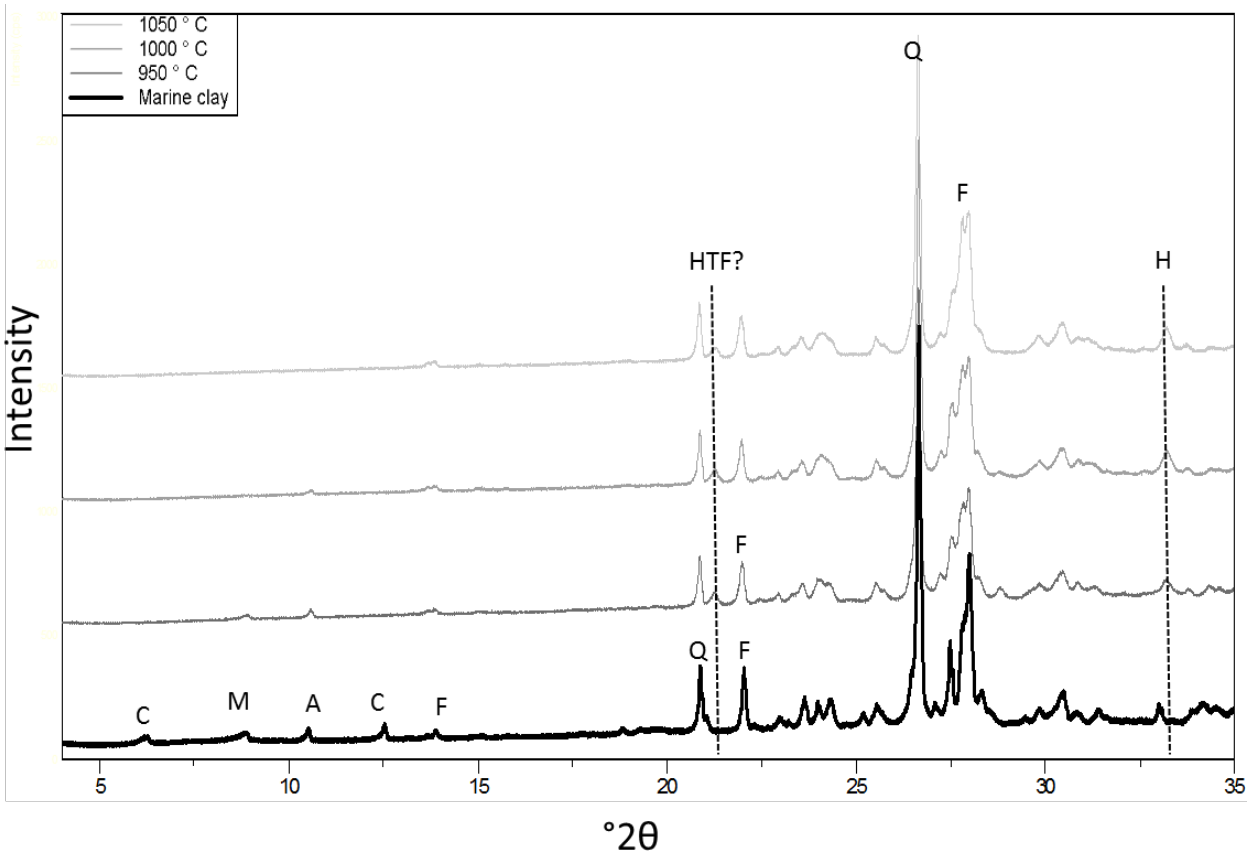

Figure 3. X-ray diffractograms of the marine clay and the pellets at the three firing temperatures: 950°C, 1000°C and 1050°C. A = amphibole, C = Chlorite, F = Feldspar, H = Hematite, HTF? = High temperature feldspar?, M = Mica, Q = Quartz.

4. Conclusion

The pellets fired at 950°C have very low firing shrinkage, indicative of a low degree of sintering. Furthermore mineralogical investigations indicate that only chlorite is fully decomposed at this temperature, whereas mica and amphibole are still observable although the peaks are diminished compared to the raw material. The temperature effect on the properties investigated in this study is larger in the range from 1000-1050°C than from 950-1000°C, indicating increased sintering above 1000°C. The pellets fired at 1050°C obtained acceptable properties and together with the other observations in this study, it can be concluded that the optimum firing temperature will be above 1000°C. The high degree of sintering in the Ilulissat marine clay-bricks fired at temperatures

in the range of 1045-1080°C observed by Belmonte et al. (2014), was not observed for the pellets in this study. The differences between the two studies must be assigned to differences in production method and firing programme.

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Appendix 7

Description: Conference paper. Proceedings of the conference: 9th International symposium on cold regions development (ISCORD), Yakutia, Russia, 1-5th of June 2010.

Title: Reuse of harbour sediments in the Greenlandic construction industry

REUSE OF HARBOUR SEDIMENTS IN THE GREENLANDIC CONSTRUCTION INDUSTRY

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Introduction

Harbour sediments are commonly dredged in order to maintain navigable waterways in and around many harbours all over the world. The dredging is necessary due to a constant inflow of sediments to the harbour from e.g. local rivers and streams. The dredged harbour sediments are often polluted and are not suitable to dump elsewhere on the ocean floor and therefore have to be deposited on land.

The purpose of this study is to investigate possibilities of using harbour sediments from the Greenlandic harbours as substitutes in the Greenlandic construction industry, mainly for concrete production and road construction.

Materials for use in the Greenlandic construction industry are shipped to Greenland from all over the world and reuse and use of resources already in Greenland would therefore make a huge contribution to the local community.

Sisimiut Harbour sediment

The sediment studied here was sampled from Sisimiut Harbour, West Greenland. Sisimiut Harbour consists of a marina used by smaller boats and an industrial area used by trawlers, ferries etc. (Ottosen and Villumsen, 2006)

Harbour sediment is not removed at present in Sisimiut, however, the high heavy metal concentrations of the sediment (Ottosen and Villumsen, 2006) might necessitate the initiation of sediment dredging and repositioning as hazardous waste on land in the future, due to the vulnerable arctic environment.

Ottosen *et al.* (2007) reckon that the heavy element concentration is related to grain size fraction for the Sismiut Harbour sediment. According to Ottosen *et al.* (2007) the heavy elements, such as Cu, Cd, Zn and Pb appear to be concentrated in the fine-grained fractions (< 63 µm), whereas the coarser grained fractions can be considered unproblematic in regards to heavy element concentrations.

Ottosen *et al.* (2007) suggested electrodialytic remediation for removing the high heavy element concentrations from the fine-grained sediment fractions. The coarser grained fractions can likely be used directly for other purposes in, e.g. the construction industry.

Material characterisation

The grain size distribution of the sediment was determined by wet and dry sieving and is shown in Figure 1. The sediment is defined as poorly sorted (according to Larsen *et al.*, 1995) and consists of approximately 90 % sand in the fraction 63-2000 µm.

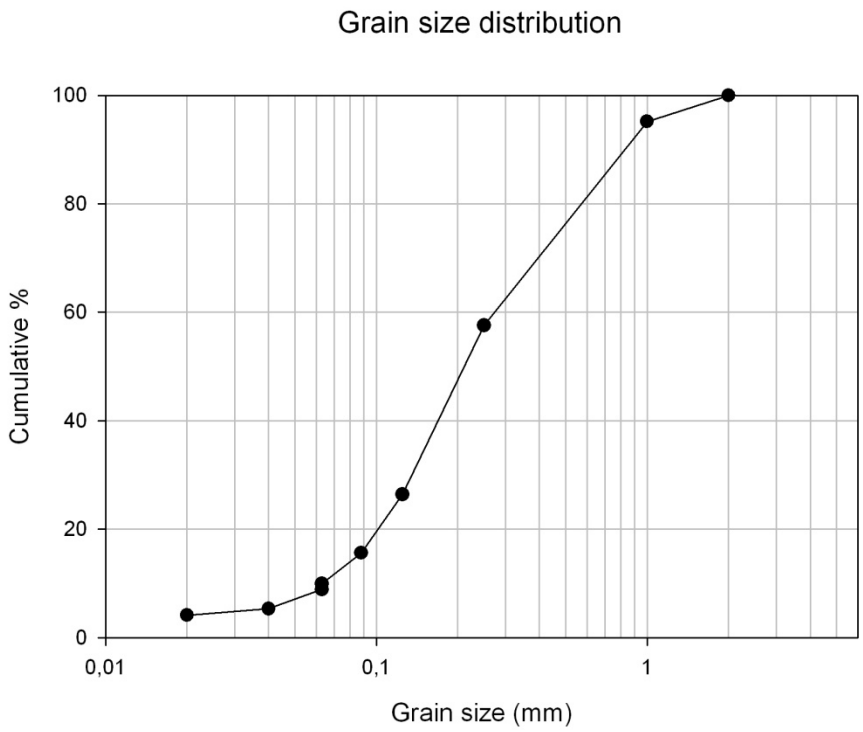


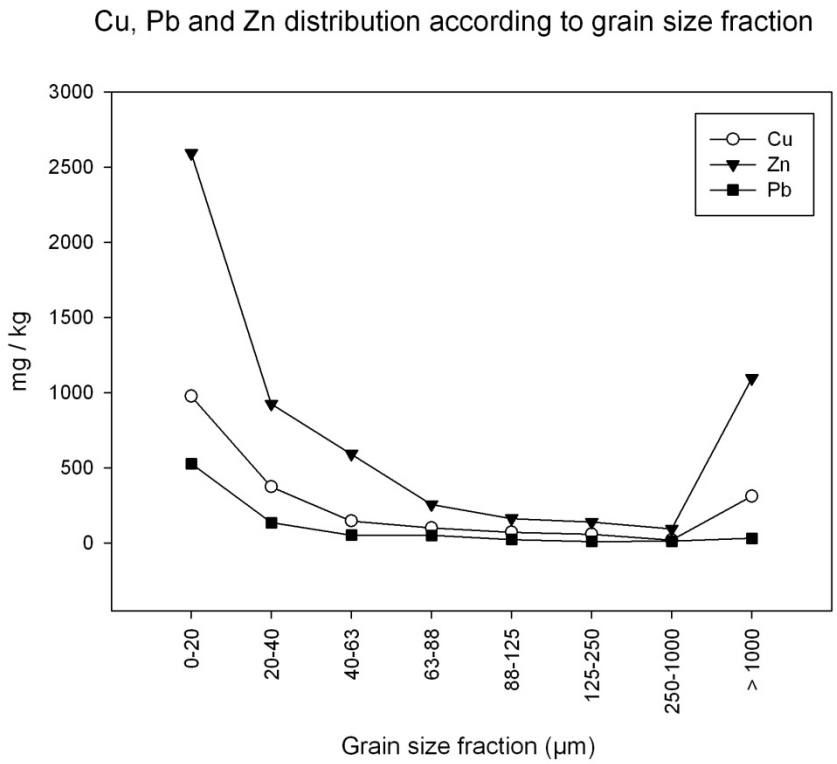
Figure 1. The grain size distribution of the Sisimiut Harbour sediment. Approximately 90 % of the sediment consists of grains in the sand fraction (63-2000 µm).

The Cu, Pb, Zn and Cd distribution in the sediment fractions: 0-20, 20-40, 40-63, 63-88, 88-125, 125-250, 250-1000 and > 1000 µm were determined after pre-treatment according to Danish Standard DS 259. According to the standard 1.0 g of dry sediment and 20.0 mL (1:1) HNO_3 were heated at 200 kPa (120°C) for 30 minutes. The liquid was separated from the solid particles by vacuum through a 45 µm nucleopore filter and diluted to 50 mL. The concentrations of the heavy metals were thereafter measured by flame or graphite furnace equipped atomic absorption spectrometry. The distribution of the heavy elements according to grain size fractions are shown in Figure 2. For all measured heavy elements the fine-grained fractions (< 63 µm) have a higher concentration than the coarser grained fractions. This is consistent with the conclusions of Ottosen *et al.* (2007).

According to the ecotoxicological assessment criteria (EAC) from the OSPAR commission, the values of Cd, Pb and Zn in grain size fraction 63-1000 µm all fall in a category where further evaluation of the sediment is needed in order to determine the risk for toxicological effects (OSPAR, 2004; Ottosen and Villumsen, 2006). The Cu values generally exceed the concentration limits recommended by the OSPAR commission, which means that the sediment is highly polluted and toxicological effects can be expected. This implies that even the coarser grained fractions of the sediment are polluted (at least by Cu) and probably need to be treated before use elsewhere.

X-ray powder diffraction (XRPD) was used to do a preliminary mineralogical characterisation of the coarser grained fractions (63-1000 µm) of the harbour sediment. The diffraction spectrum was measured with a PanAlytical X'Pert PRO diffractometer, using Cu radiation and a Ni-filter for the diffracted beam. The diffractometer is based on the Bragg-Brentano setting and has a programmable divergence slit. A voltage and current of 45 kV and 40mA were applied. The sample was measured in air for 4.5 hours. Data was treated using the PanAlytical HighScore Plus software and the PDF-2 database from the International Centre for Diffraction Data.

The sediment consisted primarily of quartz and plagioclase feldspar (Ca-rich albite) including other components, such as minor amounts of salt and possibly heavy metal containing phases.



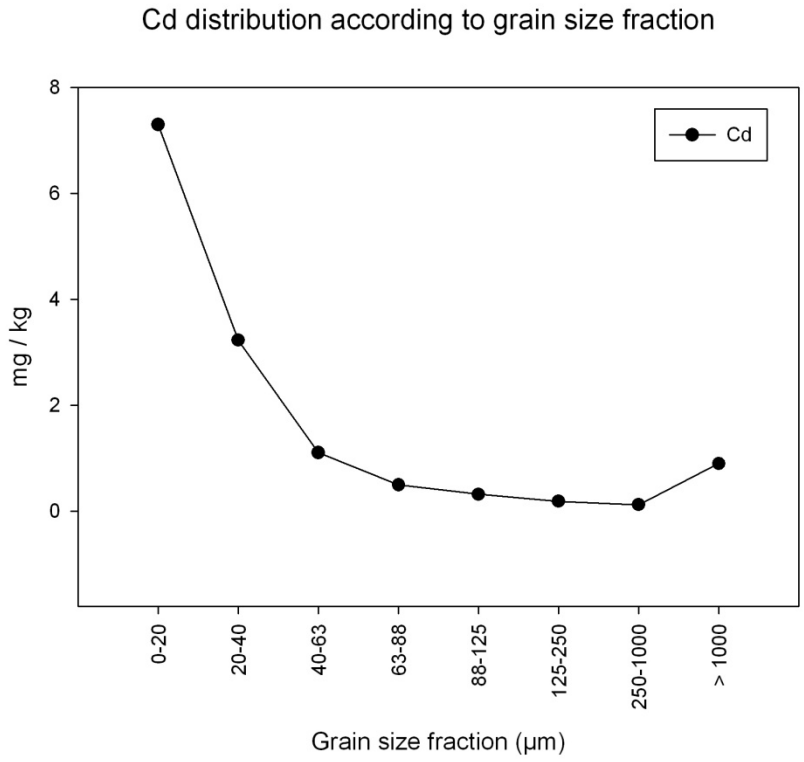


Figure 2. Cu, Pb, Zn and Cd distribution according to grain size fractions in harbour sediment from Sisimiut, West Greenland. The heavy elements are primarily concentrated in the fine-grained fractions (< 63 µm).

Aside from high heavy metal concentrations it is also likely that the harbour sediment will contain oil residues. The oil content will be investigated by gas chromatography studies.

Possible uses in the construction industry

It is likely that the coarse grained fraction (63-1000 µm) of the Sisimiut Harbour sediment can be used for geotechnical purposes, such as soil stabilizing and land filling in connection with road construction. Seen from a road construction perspective, the sediment is relatively fine-grained as it does not contain any fractions larger than 1000 µm. It can therefore be used as an additive in e.g. subbase layers where it will be mixed with other sediments and thereby create an overall well-graded material for construction.

At present there are plans to build a road from Sisimiut to Kangerlussuaq. Kangerlussuaq is located approximately 130 km W of Sisimiut and the construction of this road would therefore require vast amounts of various types of raw materials; however, further investigations, i.e. mixing of the sediment with other suitable sediments and testing of the combined bearing capacity and response to freeze-thaw cycles, would be needed to determine whether the Harbour sediment from Sisimiut could be of any use.

Whether or not the pollution of the sediment by heavy elements and possibly oil residues will prevent its use in the construction industry will also have to be investigated further; however, the heavy metals and residual oil could possibly be removed by electrodialytical remediation as suggested by Ottosen *et al.* (2007) or washed out from the sediment by a technique such as the Sediment Washing Technology developed by BioGenesis (Wilde, C., 2006).

Conclusions

The coarser grained fraction of the Sisimiut Harbour sediment consists primarily of quartz-feldspar sand and can likely be used in the construction industry as an additive for road construction. Preliminary studies have, however, demonstrated that even the coarser grained fractions of the sediment are polluted by heavy elements and the sediment might therefore have to be treated prior to further use.

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The housing situation in Greenland is critical. In 2012, approximately 20 % of the housing was dilapidated, and due to an increasing population in the larger towns, there is currently an urgent need for the construction of new homes. Today, materials used in the Greenlandic construction industry are mostly imported, and due to high import costs, it is therefore important to investigate the potential for local construction material production. The aim of this PhD-study was to identify and test suitable raw materials for a local production of clay-based bricks.

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